Synopsis of Research

Conducted under the 1999-2000 Northern Contaminants Program

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The views, conclusions and recommendations expressed herein are those of the authors and not necessarily those of the Department.

Dedication

This Northern Contaminants Program synopsis report is dedicated to Dr. H. Stuart Innes and Dr. Malcolm Ramsey, two prominent Canadian Arctic researchers whose lives were lost as a result of a tragic helicopter accident, on May 21, 2000, near Resolute Bay in Nunavut, Canada.

Dr. H. Stuart Innes, a Department of Fisheries and Oceans research scientist from the Freshwater Institute in Winnipeg, and University of Saskatchewan professor, Dr. Malcolm Ramsay, had been tagging polar bears in Barrow Strait in an effort to study the effects of global warming on polar bear and ringed seal interaction. During their return to Resolute Bay, their helicopter went down without warning into the pack ice. Both scientists were killed instantly.

With many years of work conducted under harsh arctic climatic conditions, Drs. Innes and Ramsey were particularly experienced Arctic researchers. Both shared a respect and admiration of the Arctic and its peoples and devoted their lives to advancing northern research. Their scientific expertise was internationally recognized and respected, contributing significantly to our current understanding of the mechanisms that determine population abundance and distribution of Arctic wildlife.

It is inconceivable that the lives of our two fellow scientists, and good friends, ended so abruptly. Drs. Innes and Ramsey will be deeply missed by all, and memories of their work and comradeship always remembered.

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Foreword

This report summarizes the results of research and monitoring studies on contaminants in northern Canada, and related education, communications and policy activities. These projects were conducted under the auspices of the Northern Contaminants Program (NCP). The projects cover all aspects of northern contaminants issues, as outlined in the NCP blueprints, including human health, monitoring the health of Arctic peoples and ecosystems and the effectiveness of international controls (abiotic monitoring and modelling, biotic monitoring, local contaminants concerns), education and communications, international policy and program management.

These projects were evaluated by external peer reviewers, technical review teams, a social/cultural review team, territorial contaminants committees and the NCP Management Committee to ensure that they supported the overall NCP objectives.

Further information about the NCP is available on the NCP Web site at www.inac.gc.ca/ncp .

Préface

Ce rapport résume les résultats de recherches portant sur les contaminants, d'études sur la surveillance des contaminants dans le Nord canadien, et d'activités connexes au sujet de l'éducation, de la communication et de la politique. Ces études ont été menées dans le cadre du Programme de lutte contre les contaminants dans le Nord (PLCN). Ces projets, tels que décrit dans les plans directeurs liés au programme, représentent tous les aspects portant sur les contaminants, incluant la santé humaine, la surveillance de la santé des habitants et des écosystèmes de l'Arctique et de l'efficacité des mesures de contrôle internationales (surveillance et modélisation milieux abiotiques, surveillance — milieux biotiques, préoccupations locales concernant les contaminants), l'éducation et la communication, la politique internationale et la gestion des programmes.

Ces projets ont été examinés par une évaluation par des pairs, des comités d'examen technique, un comité d'examen social et culturel, les comités territoriales sur les contaminants environnementaux, et le comité de gestion de la PLCN afin de s'assurer qu'ils répondent à l'ensemble des objectifs du programme de lutte contre les contaminants dans le Nord.

Pour de plus amples renseignements au sujet du programme de lutte contre les contaminants dans le Nord, visitez le site Web du PLCN au www.inac.gc.ca/ncp.

Introduction

The Northern Contaminants Program (NCP) was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in fish and wildlife species that are important to the traditional diets of northern Aboriginal peoples. Early studies indicated that there was a wide spectrum of substances – persistent organic pollutants (POPs), heavy metals and radionuclides – many of which had no Arctic or Canadian sources, but which were, nevertheless, reaching unexpectedly high levels in the Arctic ecosystem. The Program's key objective is *to reduce and, where possible, eliminate contaminants in northern traditionally harvested (country) foods while providing information that assists informed decision making by individuals and communities in their food use.*

Under the first phase of the NCP (NCP-I), research was focussed on gathering the data required to determine the levels, geographic extent, and source of contaminants in the northern atmosphere, environment and its people, and the probable duration of the problem. The data enabled us to understand the spatial patterns and temporal trends of contaminants in the North, and confirmed our suspicions that the major sources of contaminants were other countries. The data, which included information on the benefits from continued consumption of traditional foods, were also used to carry out assessments of human health risks resulting from contaminants in those foods. Results generated through NCP-I are synthesized in the *Canadian Arctic Contaminants Assessment Report*.

Extensive consultations were conducted in 1997-1998 to find the common elements between the concerns and priorities of northern communities and the scientific needs identified as critical for addressing the issue of contamination in Canada's North. As a result, priorities for future research are based on an understanding of the species that are most relevant for human exposure to contaminants in the North, and geographic locations and populations that are most at risk.

In 1998-1999, the NCP began its second phase (NCP-II), which will continue until 2002-2003. NCP-II supports research designed to answer questions about the impacts and risks to human health that may result from current levels of contamination in key Arctic food species. To ensure a balanced assessment of the risks, an emphasis is placed on characterizing and quantifying the benefits associated with traditional diets. Communications activities are also emphasized and supported under NCP-II. Under the leadership of the northern Aboriginal organizations, the dialogue between Northerners and the scientific community, which was initiated in NCP-I, continues to build awareness and an understanding of contaminants issues, and helps to support the ability to deal with specific contaminant issues at the local level.

In addition, the NCP effort to achieve international controls of contaminants will remain strong in NCP-II. The legally binding Long-Range Transboundary Air Pollution protocol has now been successfully negotiated and was signed by 34 countries (including Canada) at the United Nations Economic Commission for Europe Ministerial Conference in Aarhus, Denmark, in June 1998. Canada ratified this agreement in December 1998. Negotiations for a legally binding global instrument on POPs under the United Nations Environment Programme have begun, a new phase of the circumpolar Arctic Monitoring and Assessment Programme is under way, and the Canada/Russia Program on Scientific and Technical Cooperation in the Arctic and the North is continuing. NCP-II will continue to generate the data that allows Canada to play a leading role in these initiatives.

The NCP is directed by a management committee that is chaired by the Department of Indian Affairs and Northern Development, and which includes representatives from the five northern Aboriginal organizations (Council for Yukon First Nations, Dene Nation, Métis Nation-Northwest Territories, Inuit Tapirisat of Canada and Inuit Circumpolar

Conference), the Yukon, Northwest Territories and Nunavut Territorial Governments, Nunavik, and four federal departments (Environment, Fisheries and Oceans, Health, and Indian Affairs and Northern Development). The management committee is responsible for establishing NCP policy and research priorities and for final decisions on the allocation of funds. Three territorial contaminants committees in the Yukon, Northwest Territories and Nunavut (established in 1999-2000) support this national committee. Funding for the NCP-II's $5.4 million annual research budget comes from the Treasury Board and the four participating federal departments.

In 1998, the NCP Management Committee redesigned the NCP-Phase II for application under the 1999-2000 funding year. The two main initiatives undertaken were: i) the development of blueprints that represent the long-term vision and strategic direction for NCP-II; and ii) the implementation of a more open and transparent proposal review process. This new management structure is designed to ensure that the NCP remains scientifically defensible and socio-culturally aware, while at the same time, achieving real progress in terms of the Program's broad policy objectives.

Blueprints were developed for each of the four main NCP subprograms: i) Human Health, ii) Monitoring the Health of Arctic People and Ecosystems and the Effectiveness of International Controls, iii) International Policy, and iv) Education and Communications. The blueprints are used to provide the necessary guidance to project proponents for the development of proposals as well as to peer reviewers, review teams and the NCP Management Committee for evaluating proposals. They are evolving documents that are reviewed at least annually.

Under a revamped proposal review process, the NCP Technical Committee was been replaced with an external peer review process facilitated by review teams. The review of proposals is a two-pronged approach involving a scientific review by external peer reviewers, facilitated by technical review teams, and a socio-cultural review facilitated by a review team and the three Territorial Contaminants Committees (TCCs). Both sets of recommendations are considered by the Management Committee in making final funding decisions. Proposals submitted under the Education and Communications subprogram are evaluated by a review team and the three TCCs. All peer reviewers, review teams and TCCs use evaluation criteria and the blueprints to review and rate proposals. Written consent from the appropriate northern community authority or national-level Aboriginal organization is required for all projects involving field work in the North and/or analyses of samples as a condition of approval for funding. The Social/Cultural Review Team ensures that each applicable proposal has a written consent form attached and sign by the applicable authority.

This report provides a summary of the research and activities funded by the NC Program in 1999-2000, the second year of NCP-II. It is a compilation of reports submitted by project teams, emphasizing the results of research and related activities that took place during the 1999-2000 fiscal year. The report is divided into chapters that reflect the broad scope of the NCP: Human Health; Monitoring the Health of Arctic People and Ecosystems and the Effectiveness of International Controls (including abiotic monitoring, biotic monitoring, and local contaminants concerns), Education and Communications, International Policy, and Program Management. Projects that are reported in this Synopsis of Research will be presented at the 10th Annual Northern Contaminants Program Results Workshop, to be held in Calgary, Alberta, September 26-28, 2000.

Human Health

A Study with Cynomolgus Monkeys (*Macaca fascicularis*) to Determine the Potential Toxicological and Reproductive Effects of Ingesting Technical Grade Toxaphene

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Abstract

Forty adult female monkeys were randomly distributed into four dose groups (A,B,C,D) receiving 0.0, 0.1, 0.4 and 0.8 $mg{\cdot}kg^{-1}$ body $weight{\cdot}day^{-1}$, respectively, of technical grade toxaphene. Five adult male monkeys also received a dose of 0.8 $mg{\cdot}kg^{-1}$ body $weight{\cdot}day^{-1}$ and five adult males served as controls. After 75 weeks on test, during which time the females underwent immunological evaluation, the dams were mated with the untreated males over a period of 36 weeks. Concurrently, the treated males received daily doses of toxaphene for 98 weeks, at which time their dosing was stopped and the half-life of toxaphene was determined prior to necropsy. The infants were weaned at 22 weeks of age and underwent immunological evaluation prior to receiving the same daily dose of toxaphene as their dam. After 50 weeks of dosing, the infants were necropsied. The non-impregnated females were used to determine toxaphene half-lives prior to their necropsy. The dams were necropsied approximately 10 weeks after their infants were weaned. Only data pertaining to the first 75 weeks have been completely analyzed to date. These data show that there was no effect of treatment upon body weight gain, feed consumption, water consumption, haematological parameters or menstrual status. Toxaphene did affect serum cholesterol (i.e. decreasing as the dose of toxaphene increased) and resulted in the following clinical signs: a slight increase in nail bed prominence, a slight edema of the eyelids and a low incidence of dry skin.

Objectives

Long-term

- A large monkey study was initiated in May 1996 to thoroughly assess the toxicological effects of toxaphene ingestion upon the adult monkey, including reproductive effects, and the neonatal and postnatal development of the infant. The data from this study will assist in the assessment of toxaphene's potential risk to human health and will provide information to determine whether the tolerable daily intake for toxaphene, as set by Health Canada, is acceptable or warrants a review.

Short-term

1. Complete necropsy of non-depletion (impregnated) females (April 1999).
2. Complete 25-week female depletion study and necropsy depletion (non-impregnated) females (May 1999).
3. Initiate and complete immunological testing of infant monkeys (February 2000).
4. Complete necropsy of infants (March 2000).
5. Begin histopathological evaluation of necropsy samples.

Introduction

Toxaphene, a mixture of polychlorinated monoterpenes, was initially sold by Hercules Co., as Hercules 3956, in 1945. It was the most heavily used pesticide in the United States and many parts of the world prior to being banned (Voldner and Schroeder 1990, Saleh 1991). It was registered for use on more than 168 agriculture commodities and crops in the U.S. It was also used to control ectoparasites on cattle and sheep in addition to its use for fish eradication programs (Korte et al. 1979). However, it has been reported that 65% to 90% of all the toxaphene used in the United States was applied to cotton in the mid-1970s (von Rumker et al. 1974, Eichers et al. 1978, International Agency for Research on Cancer 1979, Korte et al. 1979, Federal Register 1982). Most registered usages of toxaphene in Canada were cancelled in 1980 (Agriculture Canada 1980).

At the time toxaphene was banned, it was well known that the prevailing winds had transported toxaphene for long distances (Chemical & Engineering News 1982). The first documented evidence in 1975 of this phenomenon has been attributed to Bidleman and Olney (U.S. Environmental Protection Agency 1982). Within a few years, toxaphene residues were found consistently in samples where toxaphene was either never used or not used to any significant extent (Jansson et al. 1979, Zell and Ballschmiter 1980, Sundström 1981, Schmitt et al. 1983). Toxaphene has been found in peat bogs from the Great Lakes area and through northeastern Canada, even though the predominant use of toxaphene was in the southern United States (Rapaport and Eisenreich 1986). However, whether the toxaphene found in the various Arctic samples is due to volatilization of North American soil residues or by transport from other sources is an open question (Andersson et al. 1988, Bidleman et al. 1990), although some have suggested that non-North American sources may be important contributors to the amount of toxaphene found in the Canadian Arctic (Rahn 1981, Barrie 1986, Muir et al. 1990).

The implications of the above observations were evident in a report of the mean concentration of organochlorines in the milk fat samples obtained from Inuit people living in Arctic Quebec, which were two to 10 times greater than in similar samples collected in southern Quebec (Dewailly et al. 1993). Subsequently, it was reported that 50% of the dietary intake recalls collected from an Inuit population on Baffin Island in the eastern Arctic exceeded the acceptable daily intake for toxaphene (Kuhnlein et al. 1995). Because of these findings, and the general lack of toxicological data regarding toxaphene that conforms to present standards for toxicological testing, a chronic toxicity and reproductive study was undertaken.

Forty adult female monkeys were randomly distributed into four dose groups (A,B,C,D) receiving 0.0, 0.1, 0.4 and 0.8 mg·kg^{-1} body weight·day^{-1}, respectively, of technical grade toxaphene. Five adult male monkeys also received the high dose of 0.8 mg·kg^{-1} body weight·day^{-1} and five adult males served as controls.

Activities in 1999-2000

The necropsies of the non-depletion (impregnated) females were completed on April 29, study week 154. The following week, the depletion females were necropsied. Samples of blood, fat and an array of tissues were taken from each for toxaphene analysis, histological and immmunohistological evaluation, and molecular endpoints.

The infants were weaned at 22 weeks of age. Immunological testing of the infants was initiated when the infants were 25 to 27 weeks of age. Dosing of the infants started upon completion of the immunological testing, which required four weeks. The infants were then dosed daily until they were necropsied, at which time they were 77 to 79 weeks of age. As with the adults, samples of blood, fat and an array of tissues were taken from each for toxaphene analysis, histological and immunohistological evaluation, and molecular endpoints.

Results

A manuscript for the first 75 weeks of the study, the pre-mating phase, has been submitted to the journal *Food and Chemical Toxicology*. Statistical analyses of the data did not reveal any effect of treatment upon body weight gain, feed consumption, water consumption, haematological parameters or menstrual status. Toxaphene did have an effect upon the serum biochemistry parameter cholesterol, wherein the level of cholesterol in the serum decreased with increasing dose of toxaphene. The following clinical signs were noted: a slight increase in nail bed prominence, a slight edema of the eyelids and a low incidence of dry skin in the highest dose group. However, all of these clinical signs are characteristic of halogenated hydrocarbon ingestion.

Subsequent manuscripts require statistical, histological and immunohistological evaluation, molecular endpoints and/or chemical analyses of tissues before they can be completed. In addition, the analyses of the infant immunological data has not yet been completed. Once completed they will be submitted for statistical evaluation.

Discussion

For the clinical signs, there did not appear to be any real dose relationship, save for the incidence of dry skin primarily observed at the high dose level.

Preliminary analyses of the in utero and neonatal growth data indicate that the female fetuses and neonates of the toxaphene treated dams increased in size at a faster rate than did the fetuses and neonates of the control dams.

The time for the completion of several aspects of subsequent manuscripts will depend upon the availability of statistical, analytical and pathological resources.

Project Completion Date

In vivo portion of study was completed in March 2000. Further analyses will continue subject to available resources.

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Adverse Developmental Effects in Pigs Following in Utero and Lactational Exposure to Organochlorines: Effects on the Immune System

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Abstract

This study was set up to investigate adverse developmental effects resulting from in utero and lactational exposure to an environmentally relevant mixture of organochlorines (similar to that found in the blubber of northern Quebec ringed seals consumed by the Inuit population), using the pig as the animal model. Immune function parameters (vaccination response, lymphocyte subsets, complement system component) were measured at various times during the first year of life in F1 males whose sows were exposed to various doses of the organochlorine mixture or to the vehicle alone (corn oil). There were no treatment-related effects on the percentage of lymphocyte subsets (CD4+, CD8+, CD4+CD8+, SLA-DR, CD8+DR) or C2-component mediated haemolytic activity. However, a lower proportion of animals in the high-dose group (100 μg PCB·kg^{-1} body weight) had *Mycoplasma hyopneumoniae* antibody titers ≥ 7 mg·L^{-1} at 6, 7 and 8 months of age, compared to the other experimental groups. These results indicate that prenatal and early postnatal exposure to organochlorines alter the immune system response following vaccination. Vaccination response is presently being assessed in the Inuit population in relation to perinatal organochlorine exposure.

Objectives

- To investigate the possible relationship between exposure to an organochlorine mixture similar to that found in the Arctic aquatic food chain and various adverse developmental endpoints. The present report focuses on immunological effects.

Introduction

Organochlorine compounds can be found in virtually all ecosystems of the planet. This group of chemicals includes pesticides (e.g. dieldrin, mirex, toxaphene), industrial compounds and by-products of various industrial processes (e.g. HCB [hexachlorobenzene], PCBs [polychlorinated biphenyls], PCDDs [polychlorodibenzo-*p*-dioxins] and PCDFs [polychlorodibenzofurans]). These compounds are biomagnified in food chains, and human populations relying on fish or marine mammals for sustenance receive an unusually high dose of organochlorine compounds through their diet (Dewailly et al. 1993).

Several organochlorine compounds display immunotoxic properties in both laboratory animals and humans, the most potent being substances structurally related to TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin) such as non- and mono-ortho chloro-substituted PCBs, as well as 2,3,7,8-chloro-substituted PCDDs and PCDFs. In almost all animal species tested, including primates, PCDDs, PCDFs and PCBs produce myelosuppression, immunosuppression, thymic atrophy and inhibition of immune complement system components (National Research Council 1992). Evidence in laboratory animals suggests that the maturation of the immune system is especially vulnerable to the adverse effects of dioxin-like compounds, chlordane, polycyclic aromatic hydrocarbons and possibly endocrine disrupting compounds such as DDT and kepone (Holladay and Luster 1996).

Infants born to accidentally exposed Yu-Cheng mothers had more episodes of bronchitis or pneumonia during their first 6 months of life than unexposed infants from the same neighborhood (Rogan et al. 1988). The authors speculated that the increased frequency of pulmonary diseases could result from a generalized immune disorder induced by transplacental or breast milk exposure to dioxin-like compounds, more likely PCDFs (Rogan et al. 1988).

We recently reported that the risk of otitis media in Inuit infants from Nunavik (northern Quebec) was associated with prenatal exposure to organochlorine compounds (Dewailly et al. 2000). This population is exposed to a complex mixture of organochlorine compounds and heavy metals through their traditional diet, which includes sea mammal tissues. The present study investigates changes in immune function following transplacental and lactational exposure to a mixture of organochlorine compounds similar to that found in the Arctic aquatic food chain, using the pig as the animal model.

Activities in 1999-2000

Animal treatment

Sixteen Landrace-Yorkshire-Duroc sows were randomly allocated to four treatment groups. Animals in each group were administered different doses of an organochlorine mixture from 4 months of age until weaning of their first litter. The composition of the organochlorine mixture is described in Table 1 and was designed to approximate that found in the blubber of ringed seal from Nunavik (Derek Muir, personal communication). Organochlorines were dissolved in corn oil to reach the appropriate concentration and the resulting solution was placed in 2-mL gelatine capsules. The first group received 1 µg, the second group 10 µg and the third group 100 µg PCB·kg^{-1} body weight·day^{-1}. Animals in the fourth group (control group) were administered corn oil only. At puberty, females were inseminated with semen from an untreated Duroc boar. Eight to 10 male piglets per dose group were nursed by their sow for 3 weeks and received a standard diet after weaning. Piglets were vaccinated against *Mycoplasma hyopneumoniae* at 6 weeks of age. Blood samples were collected monthly for organochlorine determination and for immunological measurements.

Table 1. Composition of the organochlorine mixture

Compound	CAS* number	% weight
PCB mixture**		32.59
technical chlordane	57-74-9	21.30
p,p'-DDE	72-55-9	19.24
p,p'-DDT	50-29-3	6.79
technical toxaphene	8001-35-2	6.54
α-HCH	319-84-6	6.17
aldrin	309-00-2	2.52
dieldrin	60-57-1	2.09
1,2,4,5-tetrachlorobenzene	95-94-3	0.86
p,p'-DDD	72-54-8	0.49
β-HCH	319-85-7	0.46
hexachlorobenzene	118-74-1	0.35
mirex	2385-85-5	0.23
γ-HCH	58-89-9	0.20
pentachlorobenzene	608-93-5	0.18

* Chemical Abstract Service

** PCB mixture containing 2,4,4'-trichlorobiphenyl (320 mg), 2,2',4,4'-tetrachlorobiphenyl (256 mg), 3,3',4,4'-tetrachlorobiphenyl (1.4 mg), 3,3',4,4', 5-pentachlorobiphenyl (6.7 mg), Aroclor 1254 (12.8 g) and Aroclor 1260 (19.2 g)

Blood sampling and processing

Blood samples were collected in vacutainers containing heparin as the anticoagulant. Samples were immediately processed for lymphocyte isolation using a Ficoll-Hypaque gradient. Plasma was transferred in glass vials pre-washed with hexane and stored frozen at −20°C. Plasma samples were sent to the Centre de Toxicologie du Québec for organochlorine determination.

Organochlorine analysis

A 1:1:3 mixture of ammonium sulfate, ethanol and hexane was first added to the plasma for organochlorine extraction. Extracts were then concentrated and purified on Florisil columns. Fourteen PCB congeners, chlorinated pesticides and their metabolites (Table 2) were measured in purified extracts by high-resolution dual-capillary column (Hewlett-Packard Ultra I and Ultra II) gas chromatography with dual ^{63}Ni electron capture detectors. Detection limit for most compounds was 0.02 $\mu g \cdot L^{-1}$.

Immune system parameters

Vaccination response: monthly antibody titers (*Mycoplasma hyopneumoniae*) were determined by an ELISA method (BIOVET, Saint-Hyacinthe, Quebec).

Phagocytosis: polymorphonuclear cells were incubated with increasing concentrations of fluorescent bacterial particles and the number of functional cells were determined by flow cytometry using the Vybrant Phagocytosis Assay Kit (Molecular Probes) (1, 5, 8 months).

Lymphocytes surface markers: cells were stained with the appropriate FITC-conjugated mAb markers and analyzed by flow cytometry for determination of the following subsets: CD4+, CD8+, CD4+CD8+, SLA-DR and CD8+DR (1, 2, 4, 6 and 8 months).

Complement system: C2 component activity was determined in plasma samples collected at 1, 5 and 7 months by an haemolytic complement assay.

Results

Weight gain of sows during treatment was similar in all groups (data not shown) and there were no signs of toxicity. Table 2 presents mean concentrations of the most abundant organochlorines in plasma samples collected after 30 weeks of treatment. Organochlorine compounds were not found in plasma samples from sows belonging to the control group, with the exception *p,p'* -DDE which was present in concentrations near the detection limit. Concentrations of the main organochlorine compounds in samples from sows in the 100 $\mu g \cdot kg^{-1}$ dose group are seven- to 10- fold greater than those measured in samples from animals in the 10 $\mu g \cdot kg^{-1}$ group. Only the most abundant and persistent compounds were detected in plasma samples from the low-dose group (1 $\mu g \cdot kg^{-1}$ group).

There was no statistically significant difference between groups with regard to birth weight, length or ano-genital distance of male piglets (data not shown). No difference between groups was observed for phagocytosis, surface markers or C2 component activity measurements (data

Table 2. Concentration of organochlorines ($\mu g \cdot L^{-1}$) in plasma samples collected from sows treated for 30 weeks with the organochlorine mixture and from control animals

	Controls*	1 $\mu g \cdot kg^{-1}$ body weight*	10 $\mu g \cdot kg^{-1}$ body weight*	100 $\mu g \cdot kg^{-1}$ body weight*
β-HCH	ND**	ND	0.05 ± 0.01	0.30 ± 0.02
p,p'-DDE	0.07 ± 0.06	0.21 ± 0.03	2.40 ± 0.31	19.0 ± 2.2
p,p'-DDT	ND	0.07 ± 0.01	0.68 ± 0.07	4.3 ± 0.2
hexachlorobenzene	ND	ND	0.07 ± 0.02	0.58 ± 0.10
mirex	ND	ND	0.06 ± 0.01	0.43 ± 0.04
oxychlordane	ND	ND	0.07 ± 0.01	0.62 ± 0.02
trans-nonachlor	ND	ND	0.11 ± 0.04	0.55 ± 0.10
α-HCH	ND	0.03 ± 0.01	0.27 ± 0.09	1.80 ± 0.20
dieldrin	ND	ND	0.22 ± 0.09	1.20 ± 0.12
ΣPCBs***	ND	0.12 ± 0.03	2.03 ± 0.26	14.4 ± 0.6

* Arithmetic mean ± standard deviation

** Not detected

***Sum of 14 PCB congeners (International Union of Pure and Applied Chemistry numbers 28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187) not shown). Table 3 shows the percentage of male piglets with plasma antibody levels against *Mycoplasma hyopnemoniae* ≥ 7 mg·L^{-1}. Exposure to organochlorine compounds did not modify substantially the percentage of animals responding to the vaccine during the first months of life. However, at 6, 7 and 8 months of age, a lower percentage of animals in the high-dose group (100 μg·kg^{-1}) reached a 7 mg·L^{-1} antibody concentration (seroconversion level) compared to the other groups. For example, at 8 months of age, only 10% (1/10) of animals in the high-dose group were above this level, compared to 80% (8/10) in the middle dose group ($p = 0.005$; Fisher's exact test), 70% (7/10) in the low dose group ($p = 0.02$) and 50% (3/6) in the control group ($p = 0.12$).

Discussion and Conclusions

These results indicate that the high dose of the organochlorine mixture can interfere with immune system function in this animal model, as suggested by the low percentage of animals with proper antibody response following vaccination. Total PCB concentration in maternal plasma during gestation reached approximately 20 mg·kg^{-1} lipids, a concentration approximately 10-fold higher than previously documented in Inuit women (Dewailly et al. 1993). Studies are currently underway to investigate the efficacy of vaccination programs currently implemented in the Inuit population of Nunavik.

Project Completion Date

The project was completed on March 31, 2000.

Acknowledgments

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Table 3. Percentage of male piglets showing plasma antibody* levels > 7 mg·L^{-1} during the first 8 months

Group/Age	1 month	2 months	3 months	4 months	5 months	6 months	7 months	8 months
Control	0	75.0	50.0	62.5	50.0	87.5	62.5	50.0
1 μg·kg^{-1} body weight	0	92.3	75.0	27.3	50.0	55.6	50.0	70.0
10 μg·kg^{-1} body weight	20.0	80.0	66.7	16.7	36.4	70.0	50.0	80.0
100 μg·kg^{-1} body weight	0	66.7	66.7	30.7	58.3	30.0	20.0	10.0

* *Mycoplasma hyopneumoniae* antibodies; vaccine administered at 6 weeks of age.

Dietary Effects on Methylmercury Toxicity in Rats

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Abstract

This ongoing project is designed to study the possible effects of dietary modulations on mercury toxicity. Mercury accumulates in fish and marine mammals; therefore, Inuit may be exposed to higher levels of mercury from their traditional diet. The traditional diet, however, also contains rich sources of beneficial nutrients such as protein, oil, vitamins and minerals. There is some evidence that these nutrients may protect against mercury toxicity. This project will study effects of these nutrients on mercury toxicity by feeding rats diets containing different nutrient compositions. The focus in 1999-2000 was on formulating and characterizing diet compositions. Experiments will begin in fall 2000. Results will be useful for characterization of the health risk associated with mercury in the Inuit diet.

Objectives

Overall objective:

- To study the effects of dietary factors on methylmercury (MeHg) toxicity.

Specific objectives:

1. To study the effects of ringed seal oil on MeHg kinetics and toxicity.
2. To study the effects of selenium on MeHg kinetics and toxicity.
3. To study the effects omega-3 fatty acids on MeHg kinetics and toxicity.
4. To study the effects of vitamin E on MeHg kinetics and toxicity.
5. To study the combined effects of ringed seal oil, selenium and vitamin E on MeHg kinetics and toxicity.

Introduction

Elevated levels of mercury (Hg) have been reported in the terrestrial, freshwater and marine biota in the Canadian North (Jensen et al. 1997). Ringed seal (*Phoca hispida*), the most abundant marine mammal species in the Canadian Arctic, is a major component and the major source of Hg in the Inuit traditional diet (Chan et al. 1995). Elevated levels of Hg, i.e. exceeding the Health Canada consumer guideline (for fish flesh) of 0.5 $\mu g \cdot g^{-1}$ wet wt, are commonly found in ringed seal meat (Chan 1998). Results of the Broughton Island study in 1988 showed that the Hg intake levels of 83% of the respondents exceeded the Provisional Tolerable Weekly Intake of 5 $\mu g \cdot kg^{-1} \cdot week^{-1}$ (Chan et al. 1997). The Inuit diet also contains high concentrations of selenium (Se) and omega-3 fatty acids.

In the recent workshop, Scientific Issues Relevant to Assessment of Health Effects from Exposure to Methylmercury, held by the National Institute of Environmental Health Sciences in North Carolina (November 18-20, 1998), it was agreed that dietary factors may affect MeHg toxicity, but there are inadequate data.

The Northern Contaminants Program Blueprint for Human Health has also identified the importance of establishing interactions between contaminants and other beneficial substances in the diet (such as Se and omega-3 fatty acids) when formulating advice to Northerners (Northern Contaminants Program 1999).

Dietary factors influence the whole-body retention of orally administered MeHg and mercuric Hg. In mice, it was found that milk increased MeHg absorption from the gastro-intestinal tract (Landry et al. 1979). This could result from an effect of milk on the enterohepatic circulation and/or de- and remethylation of Hg in the intestine since the absorption rate of MeHg is close to 100% in all cases. Wheat bran added to semi-synthetic diets decreased, whereas cellulose increased, the retention of orally administered MeHg chloride in mice (Rowland et al. 1986). A high-protein, low-fat diet decreased the retention of orally administered MeHg in mice (Rowland et al. 1984). High levels of vitamin E decreased mortality induced by both MeHg and mercuric dichloride in Japanese quails (Ganther et al. 1974, Welsh and Soares 1976). Diets containing fish protein, a good source of both Se and sulphur amino acids, decreased the toxicity of MeHg in mice compared with diets containing casein (Ohi et al. 1976). Hojbjerg et al. (1992) found that the whole-body retention was significantly reduced when mice were fed with a diet containing 50% cod liver oil compared with mice fed with 50% coconut oil. However, the relative deposition of Hg was significantly higher in all organs of mice fed a diet containing 20% energy from cod liver oil compared with mice fed a diet containing 20% energy from soya oil.

It has been more than 30 years since Parizek and Ostadalova (1967) described the protective effect of selenite against mercuric chloride intoxication. Since then, the interaction between Hg and Se has been the subject of intensive research. The role of Se, particularly the role of biological Se, in the protection against MeHg, however, has remained unquantified or even uncertain (Magos 1991).

In support of the protective role of biological Se, diet supplemented with seafood high in Se delayed the onset of MeHg intoxication (Ganther et al. 1972, Ohi et al. 1976, Freeman et al. 1978). However, these experiments did not define the contribution of other dietary constituents present in the selected seafood. Ohi et al. (1976) found that a tuna diet was half as effective in preventing the neurological manifestations of MeHg as a selenite-supplemented casein diet that contained as much Se as the tuna diet. The bioavailability of biological Se for HgSe formation is < 20% of the bioavailability of selenite Se (Magos 1991). Moreover, Ohi et al. (1976) also found that Se is not the only constituent that offered protection; the increase of casein in the diet without Se supplement also delayed neurological manifestation. Stillings et al. (1974) also reported increased growth and survival of MeHg-exposed rats with increase in casein or fish protein in the diet.

There are other problems regarding Se protection. For example, Magos and Webb (1977) reported that selenite could temporarily increase the concentration of MeHg in the brain, which is the target organ for toxicity. They studied the effect of the selenite (5 $\mu mol \cdot kg^{-1}$) on the brain concentration of Hg in rats treated with 5 $\mu mol \cdot kg^{-1}$ MeHg. Selenite was administered at day 2, 3 and 7 after MeHg treatment, and 24-hour change in brain Hg concentration was +134%, +103% and +88%, respectively. The protective role of selenite is also dubious during pregnancy (Magos 1991). Nishikido et al. (1988) demonstrated that Se deficiency did not influence the teratogenicity of MeHg, and at a high level of MeHg exposure, selenite decreased resorption only at a dose that increased the incidence of cleft palate. Even worse, MeHg can increase the toxicity of selenite (Yonemoto et al. 1985). Since the developing fetus is a sensitive target for MeHg, these observations cast doubt on the effectiveness of Se protection.

Activities in 1999-2000

The diet compositions have been formulated and characterized for the following experiments:

Experiment 1: Effect of dietary proteins

The hypothesis of this experiment is that fish protein protects against MeHg toxicity by enhancing the glutathione (GSH) production.

Both fish protein and whey are good sources of sulfur amino acids, which can enhance GSH production, whereas casein is a poor sulfur amino acid source. In addition, fish protein is a good source of Se. In this experiment, whey protein diet will be used to control for the effect of Se.

Thirty male rats will be fed one of three isocaloric, starch-based, semi-purified diets containing 20% wt of either (i) fish protein, (ii) whey protein, or (iii) casein for 4 weeks before the dosing experiment. MeHg will be administered for 14 days by gavage at three dose levels: (i) control, (ii) 1 $mg\cdot kg^{-1}\cdot day^{-1}$, and (iii) 3 $mg\cdot kg^{-1}\cdot day^{-1}$. During the course of dosing, the rats will stay on the three semi-purified diets and the general metabolic indices will be monitored. At the end of the dosing period animals will be sacrificed, necropsied and selected tissues analyzed for Hg analysis and for toxicological and histopathological assessment. The relationship between histopathology and tissue content of Hg and GSH will be examined.

The formulated diet composition is shown in Table 1.

Experiment 2: Effect of dietary lipids

The hypothesis for this experiment is that seal oil protects against MeHg toxicity by modulating lipid peroxidation. Seal oil contains a high concentration of n-3 polyunsaturated fatty acids. Lard contains high concentrations of saturated fatty acids and corn oil contains a low concentration of n-3 fatty acids but high concentrations of n-6 polyunsaturated fatty acids.

Thirty male rats will be fed one of three isocaloric, starch-based, semi-purified diets containing 20% wt of either (i) seal oil, (ii) lard or (iii) corn oil for 4 weeks before the dosing experiment. MeHg will be administered for 14 days by gavage at three dose levels: (i) control, (ii) 1 $mg\cdot kg^{-1}\cdot day^{-1}$, and (i) 3 $mg\cdot kg^{-1}\cdot day^{-1}$. Animal treatment and tissue analyses are the same as in Experiment 1.

The composition of the basal diet was formulated as shown in Table 2.

For ease of comparison with future experiments, we decided to use oil obtained from commercial sources. Fish oil is obtained from Omega Protein (Hammond, Louisiana, U.S.A), DHA oil from Martek Corporation (Columbia, Maryland, U.S.A) and seal oil from Atlantis Marine Inc.(St. John's, Newfoundland). The peroxide values, vitamin E, Se and Hg concentrations of the oil samples were characterized (Table 3).

Results and Discussion

This ongoing project is designed to study the possible effects of dietary modulations on mercury toxicity. The focus in 1999-2000 was on formulating and characterizing diet compositions for the first two of three planned experiments (Tables 1-3).

Experiments 1 and 2 will begin in fall 2000. Following their completion, Experiment 3 (Effects of Se, vitamin E, fish protein and seal oil on Hg-induced oxidative stress

Table 1. Basal diet composition, Experiment 1: Effects of dietary proteins

Component	%	Omega Protein Silver Herring™ low ash Menhaden fish meal ($g\cdot kg^{-1}$)	ICN casein vitamin free ($g\cdot kg^{-1}$)	Avonmore whey (Provon 190) ($g\cdot kg^{-1}$)
Corn starch	45.95	404.99	441.49	439.49
Protein (fish meal or casein or whey)	20.00	260.00	218.00	220.00
Sucrose	10.0	100.00	100.00	100.00
Fiber	5.0	50.00	50.00	50.00
Fat (20% butter, 20% lard, 10% corn oil, 50% soybean oil)	14.0	140.00	140.00	140.00
Mineral mix (AIN-93G)	3.5	35.00	35.00	35.00
Vitamin mix (AIN-93-VX)	1.0	10.00	10.00	10.00
L-cystine	0.3	None (0.7% already present)	3.00	3.00
Choline bitartrate (41.1% choline)	0.25	None (1.46 g already present)	2.50	2.50
Tert-butylhyro-quinone	0.0014	0.014	0.014	0.014

Table 2. Basal diet composition, Experiment 2: Effect of dietary lipids

Ingredients	g·kg^{-1} in diet
Corn starch	431.516
Casein*	200
Safflower oil	16
Sucrose	100
Fiber	50
Oil**	150
Mineral mix (AIN-93G)	35
Vitamin mix (AIN-93G)	10
L-Cystine	3
Choline bitartrate (47.85% choline)	4.47
Tert-butylhydroquinone	0.014
Metabolizable energy	18.0 MJ·kg^{-1}

* Vitamin-free casein

** Lard, corn oil, fish oil, seal oil and DHA (40%)

and toxicity, will be conducted, with the choice of dosage to be decided based on the results of Experiments 1 and 2.

Results of this study will be useful for characterization of the health risk associated with Hg in the Inuit diet.

Expected date of completion

March 2001

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Table 3. Characterization of oils obtained from commercial sources* for use in Experiment 2: Effect of dietary lipids

	Lard	Soybean oil	Fish oil	Seal oil	DHA oil
Peroxide value	8.2	2.6	6.0	8.6	3.0
Vitamin E (mg·100g^{-1})	1.2	18.19	0.00	0.411	0.25
Se (mg·kg^{-1})	0.2	0.00	1.19	ND**	ND
Hg (mg·kg^{-1})	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2

* Lard and soybean oil from ICN pharmaceuticals (Costa Mesa, CA, U.S.A.); fish oil from Omega-protein (Hammond, LA, U.S.A.); seal oil, from Atlantis Marine Inc. (St. John's, NF); DHA oil, from Martek Corporation (Columbia, MD, USA)

** Not detected

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Toxicology of Mercury and Selenium in Ringed Seal Tissues (Year 2)

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Abstract

It has been reported that mercury and selenium interact in marine mammal tissues and may affect the toxicity of mercury. The chemical form of the mercury-selenium complex in ringed seals (*Phoca hispida*) has never been characterized. Extensive chromatographic techniques and mass spectrometry have been used to isolate and characterize the mercury and selenium ligands in seal liver. Mercury and selenium were bound to ligands within the cell membranes of ringed seal liver. The mercury-selenium binding protein has a molecular weight range of about 65 kDa, and mercury and selenium have a 1:1 molecular ratio. The mercury-selenium binding protein may contain three major polypeptides with molecular weights of 6510.8, 14 305.1 and 14 353.1 Da. The toxicology of this mercury-selenium binding protein will be studied using an animal feeding model.

Objectives

The overall objective of this project is to study the interactions between mercury (Hg) and selenium (Se) in ringed seal tissues and to elucidate the potential protection of methylmercury toxicity by Se in the traditional Inuit diet. Specific objectives of this project are to:

1. Isolate and characterize the different species of Hg and Se in muscles, livers, brain and kidneys of ringed seals.
2. Conduct an animal feeding experiment to study the toxicity of co-ingestion of the biological Hg and Se found in seal meat using a rat model.

Introduction

Elevated levels of Hg have been reported in the terrestrial, freshwater and marine biota in the Canadian North (Jensen et al. 1997). Ringed seal (*Phoca hispida*), the most abundant marine mammal species in the Canadian Arctic, is a major component and the major source of Hg in the Inuit traditional diet (Chan et al. 1995). Since a high correlation between Hg and Se has been reported in many species of marine mammals, and Se was proposed to offer protection against Hg toxicity, the interaction between Hg and Se becomes an important research subject. The participants in the Northern Contaminants Program (NCP) Mercury Workshop in December 1997 identified the study of the role of Se in Hg toxicity protection as one of the top research priorities of the NCP.

The chemical form of the Hg-Se complex in ringed seals has never been characterized. Tiemannite granules were identified in the livers of cetaceans *Ziphius cavirostris* and *Tursiops truncatus* (Martoja and Berry 1980). This Hg-Se complex, though never characterized, was suggested to be the last stage of the detoxification process through the demethylation of Hg, leading to the formation of a non-bioavailable compound. A stable Hg-selenoprotein was reported in dolphin livers (Palmisano et al. 1995, Cavalli and Cardellicchio 1995). Similarly, it was suggested that in pilot whale liver Se is involved in promoting the binding of Hg with less critical proteins after the formation of Se-trisulphide groups (Caurant et al.1996). An unstable adult of bis-methylmercury selenide has been described been in rodents (Magos 1991). It has been suggested the Hg-Se in human serum may be a heparin-binding protein (Yoneda and Suzuki 1997a, 1997b).

Activities in 1999-2000

The research effort in 1999-2000 focussed on the isolation and characterization of the Hg-Se complex in seal tissues.

Seal tissues were collected by Dr. Michael Kwan at the Nunavik Research Centre in Kuujjuaq, Quebec, and shipped on ice pack to the Centre for Indigenous Peoples' Nutrition and Environment, Ste-Anne-de-Bellevue, Quebec, for analysis. Seal livers were freeze-dried and the membrane bound Hg-Se complex was extracted using a 0.2M Phosphate buffer (pH 7.0) + 2% sodium dodecyl sulphate (SDS). Liver tissues were homogenized with a Polytron for 30 seconds, shaken for 30 minutes and centrifuged at 10 000 g at 4°C for 30 minutes. One mL of seal liver extract was eluted on Sephadex G-75 with 5 mM Tris/HCl buffer (pH 7.0) at 0.8 mL·min^{-1} and fractions were collected every 3 mL. The protein peaks were monitored with an ultraviolet (UV) detector at 280 nm. Hg and Se were measured for each fraction. Molecular weight (MW) markers, including Aprotinin (MW = 6500), lysozyme (MW = 14 300), carbonic anhydrase (MW = 29 000) and albumin (MW = 66 000), were also used to determine the molecular range of the protein peaks of seal livers. The Hg-Se complex was confirmed by high performance liquid chromatography (HPLC) coupled with inductively coupled plasma-mass spectrometry (ICP-MS). 50-100 µL of fraction containing Hg and Se was applied onto a size-exclusion-HPLC (SEC). Progel™-TSK G4000 PWXL (Supelco) column (particle size 10 µm, sample MW 200-300 × 10^3, 30 cm × 7.8 mm id) and the column was eluted with 10 mM Tris/HCl (pH 8.6) at a flow rate of 0.4 mL·min^{-1}. The eluate was introduced directly into the nebulizer tube of an ICP-MS instrument (Elan 5000 PE-SCIEX) operated under the following conditions: forward power 1000 W, plasma gas (Ar) flow rate 15 L·min^{-1}, auxilliary gas (Ar) flow rate 0.8 L·min^{-1}, nebulizer gas (Ar) flow rate 1.0 L·min^{-1}. Isotopes ^{78}Se and ^{199}Hg were monitored.

The seal liver extract was analyzed using reverse-phase (RP)-HPLC. A LC_{18} column (Supelco, 25 cm × 4.6 mm, particle size 5 µm) was used. The mobile phase was a mixture of solution A (1% trifluoroacetic acid) and solution B (acetonitrile:H_2O [60:40] plus 0.08% trifluoroacetic acid) with gradient at 0.5 mL·min^{-1}. The protein peaks were monitored with a UV detector both at 280 nm and 254 nm.

Fractions were collected according to protein peaks. Mercury and Se levels in the peaks were measured by ICP-MS. Mercury and Se concentrations in the fractions (from either RP-HPLC or Sepadex G-75 column) were measured by ICP-MS instrument (Elan 5000 PE-SCIEX) under the following conditions: forward power 1000 W, plasma gas (Ar) flow rate 15 L·min^{-1}, auxilliary gas (Ar) flow rate 0.8 L·min^{-1}, nebulizer gas (Ar) flow rate 1.0 L·min^{-1}, sample uptake rate 1.0 mL·min^{-1}. Isotopes ^{82}Se and ^{202}Hg were monitored.

Two fractions containing Hg and Se were lyophilized and further characterized by mass spectroscopy (MS). The lyophilized RP-HPLC fractions were reconstituted in 70% aqueous acetic acid and analyzed by electrospray ionization-MS (ESI-MS) using a triple quadrupole mass spectrometer (API III MS/MS system, SCIEX, Thornhill, Ontario). The solutions were infused into the electrospray ion source (fused silica capillary of 100 µm id) at a rate of 1 µL·min^{-1} from a low-pressure infusion pump (Model 22, Harvard Apparatus, South Natick, Massachusetts).

A pressurized dialysis system (Millipore Minitan System) was used to isolate the seal liver Hg-Se complex in large enough quantity for the feeding experiment (approximately 1 kg). A total of 10 kg of liver extract were fed into the system mounted with a membrane with cut-off point at 30 000 Da. The isolated protein (MW < 30 000 Da) was freeze-dried and the purity confirmed by fast performance liquid chromatography (FPLC).

Results

Highest concentrations of Hg (5.99 µg·g^{-1}) and Se (3.41 µg·g^{-1}) were found in seal livers, of which 80% were associated with cell membranes. Over 60% of Hg and Se were recovered with the Na_2HPO_4-NaH_2PO_4 and SDS extraction. A major peak that corresponded to MW

65 000 Da was found in the FPLC chromatogram (data not shown). Hg:Se molar ratios within the fraction were close to 1. The co-elution of Hg-Se in this fraction was confirmed by applying a sub-aliquot of the fraction to SEC coupled to ICP-MS. Only one protein peak was detected by both Hg and Se at 17 minutes that correspond to MW 65 000 Da (Figure 1).

The liver extract was applied to a RP-HPLC and 17 peaks were detected (Figure 2). Hg and Se were detected in two peaks labelled Peak A and B. The chemical composition of these two fractions were analyzed by ESI-MS. Peak A contained three polypeptides labelled ♦, ★, and ● had MW of 6510.8, 6608.7 and 6706.1 Da respectively (Figure 3). The MW of the polypeptides differed from each other by about 97.4 Da, which is consistent with an ionicly bound phosphate group. This indicates that the peaks labelled ★, and ● are mono- and di-phosphorylated analogues of the peak labelled ♦.

Peak B contained two polypeptides labelled ● and ✱ with MW of 14 305.1 and 14 353.1 Da, respectively (Figure 4). The two polypeptides differed form each other by 48 Da. The peaks labelled ●● and ✱✱ represent non-covalently bound phosphorylated analogues of the peaks labelled ● and ✱, with MW of 14 402.5 and 14 450.2 Da, respectively. The difference in MW between ● and ●●, or between ✱ and ✱✱ is 97.4, which is consistent with an ionicly bound phosphate group.

A total of 600 g of extract containing the Hg-Se complex was isolated. The extract contained 14.6 $\mu g \cdot g^{-1}$ total Hg, 2.02 $\mu g \cdot g^{-1}$ MeHg and 7.64 $\mu g \cdot g^{-1}$ Se.

Discussion and Conclusions

Extensive chromatographic techniques have been used to isolate and characterize the Hg and Se ligands in seal liver. Extraction of Hg and Se showed that Hg and Se were mainly bound to the cell membranes. SEC analysis result confirmed that Hg and Se had a 1:1 association and the Hg-Se binding protein had a MW range of about 65 kDa. RP-HPLC analyses found three major polypeptides associated with Hg and Se. The polypeptides had MW of 6510.8, 14 305.1 and 14 353.1 Da. It is possible these polypeptides are subunits of the Hg-Se binding protein. For example, a combination of three units of the polypeptide in Peak A (3 × 6510.8) and three units of the polypeptide in Peak B (3 × 14 402.5) will result in a protein of MW at about 62 000.

The toxicity of the isolated extract of the Hg-Se complex will be studied by a rat feeding experiment to be conducted by Health Canada.

Expected Project Completion Date

March 2001

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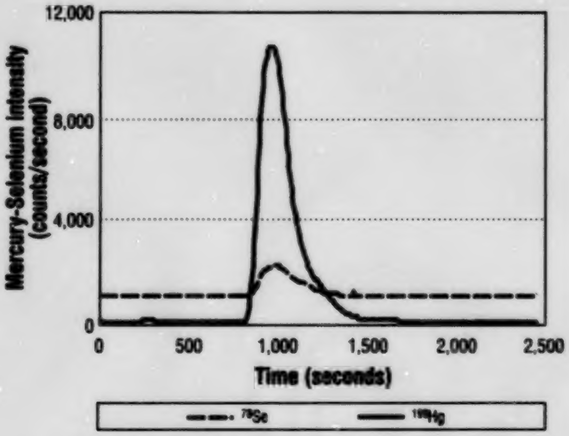
Figure 1. Size exclusion HPLC-ICP-MS chromatogram of the liver extract*



* Hg and Se co-eluted at 17 minutes

Figure 2. RP-HPLC chromatogram of the liver extract

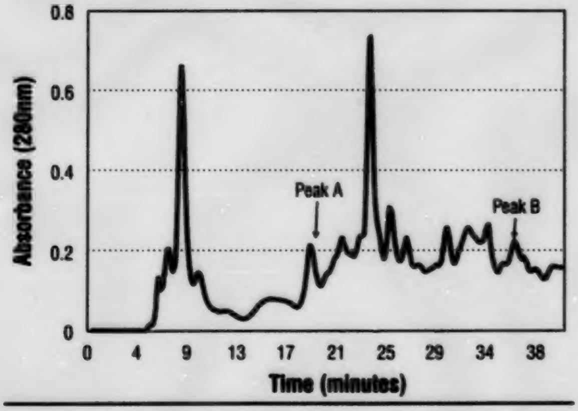
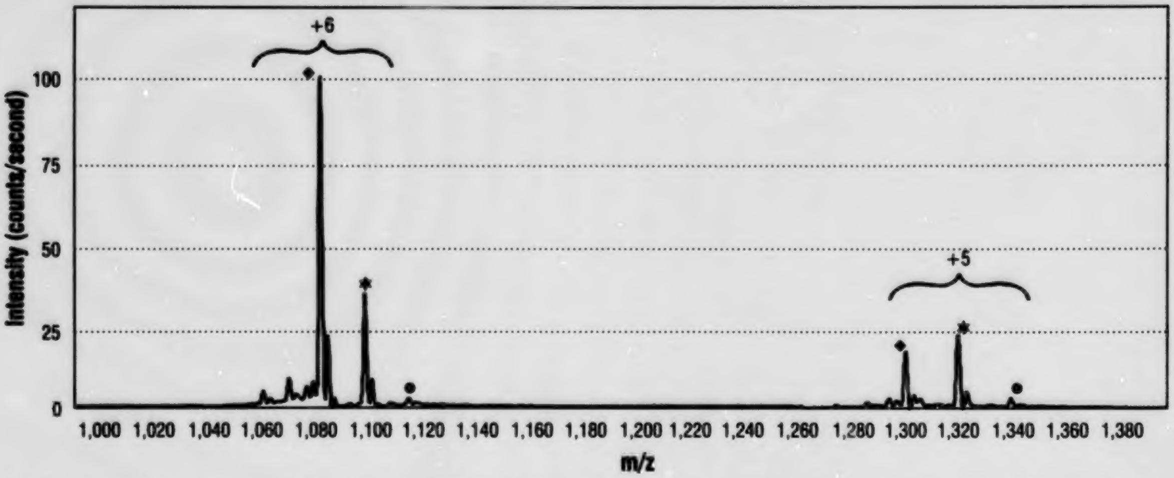
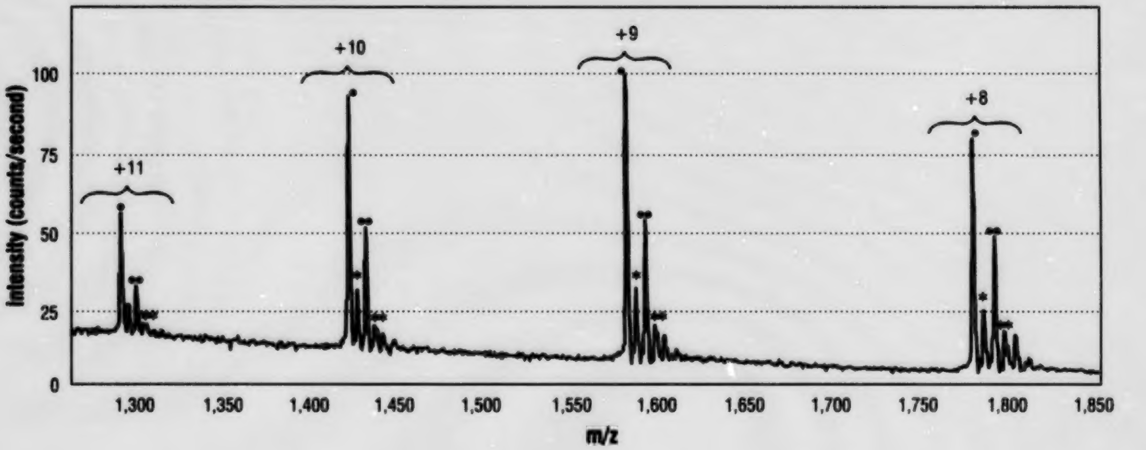


Figure 3. ESI-MS chromatogram of Peak A isolated from the RP-HPLC*



* Three polypeptides labelled ◆, ★, and ● had molecular weights of 6510.8, 6608.7 and 6706.1 Da, respectively. Peaks labelled ★, and ● are mono- and di-phosphorylated analogues of the peak labelled ◆.

Figure 4. ESI-MS chromatogram of Peak B isolated from the RP-HPLC*



* Two polypeptides labelled with ● and ✱ had molecular weights of 14 305.1 and 14 353.1 Da, respectively. Peaks labelled ●● and ✱ ✱ represent non-covalently bound phosphorylated analogues of the peaks labelled ● and ✱.

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Use of a Physiologically Based Toxicokinetic Model for Risk Assessment of Exposure to Mixtures of Organochlorines in Traditional Food (Year 3)

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Abstract

A physiologically based toxicokinetic model was developed based on the results of an animal feeding experiment. Rats were dosed with a mixture of organochlorines that were found in the Inuit traditional diet. Doses used were 10×, 100×, and 1000× of the mean estimated daily intake in humans. The rat model was scaled to a human model by substituting the model parameters with the species-specific values. Reference doses of organochlorines were estimated by determining the human exposure dose that corresponded to these liver concentrations associated with the "no observed adverse effect level" in the rat, i.e. the 1000× organochlorine concentrations. In general, the exposure doses calculated using the human physiologically based toxicokinetic model were greater than the currently used acceptable daily intakes, except for DDT and chlordane, which were similar. The recent estimation of mean daily intake of organochlorines among the Inuit communities were below the reference doses except for chlordane, which was 20 times higher than the reference dose (0.43 $\mu g \cdot kg^{-1} \cdot d^{-1}$ versus 0.02 $\mu g \cdot kg^{-1} \cdot d^{-1}$). This result suggests that the presence of chlordane in the Inuit diet may pose an increased risk to these people.

Objectives

The overall objective of this project is to develop a framework for assessing the potential health risks to the Arctic Inuit communities associated with the dietary intake of mixtures of chemical contaminants in traditional food. Specific objectives for 1999-2000 were to:

1. Scale the physiologically based toxicokinetic (PBTK) model developed for rats to humans and assess the

acceptable human exposure dose (tolerable daily intake [TDI]).

2. Assess the health risks to Inuit populations exposed to mixtures of chemicals in their diet.

Introduction

Contaminant risk assessment should ideally be based on detailed knowledge of the relationships between external and internal dose, organ levels and their relation to toxic symptoms in humans. However, human data on these toxicokinetic parameters for environmental pollutants has originated mainly from individuals or small populations accidentally exposed for short periods of time to relatively high contaminant levels, but with an unknown total body burden. Therefore, assessment of the risk associated with exposure to low levels of environmental pollutants has had to depend largely on data from animal experiments.

The use of PBTK models is a scientifically sound, mechanistic tool that has increasingly been used in health risk assessment procedures for the conduct of extrapolations essential for reducing uncertainties (Krishnan and Andersen 1994). Moreover, the recent validation of the usefulness of the physiological modelling approach in simulating the kinetics and dynamics of chemical mixtures by accounting for multiple chemical interactions (Pelekis and Krishnan 1995, Pelekis and Krishnan 1997, Tardif et al. 1997) has enabled the development of an interaction-based risk assessment approach.

Activities in 1999-2000

The first phase of this project was funded by the Northern Contaminants Program in November 1997. Phase 2 of the project was approved for funding in May 1998. This is the last phase of the project.

Based on the dietary intake data collected from Broughton Island in 1988 (Chan et al. 1997), a mixture of organochlorines was designed to contain hexachlorobenzene, α-HCH, β-HCH and δ- HCH, *oxy-*, *cis-* and *trans-*chlordane, *cis-* and *trans-*nonachlor, dieldrin, DDE, DDT, mirex, toxaphene, and PCB congeners (28, 32, 101, 99, 118, 105, 153, 128, 138, 156, 187, 183, 180, 199). Based on the toxicological dose equivalency of human studies and the 'lowest observed effect level' and 'no observed adverse effect level' (NOAEL), the dosages were decided to be 10×, 100× and 1000× the mean daily intake level. Chemicals of highest purity were purchased from a commercial supplier and the mixture was prepared in hexane at the Centre for Indigenous Peoples' Nutrition and Environment (CINE) lab and shipped to Health Canada for the feeding experiment. The mixture in hexane was added to corn oil to make up the desired dosage. Concentrations of organochlorine dose in the corn oil was confirmed by chemical analysis using gas chromatography-mass spectrometry. Four groups of 12 female rats were administered the mixture in corn oil by gavage (three dose levels plus controls) for 28 days. The animals were necropsied at the end of the experiment and organochlorine levels in liver and adipose tissue are being used to validate the mixture in the PBTK model.

Preliminary results of the toxicity test were presented in the 31st Annual Symposium of the Society of Toxicology of Canada (Suzuki et al. 1998). There were no overt signs of toxicity in any of the dose groups throughout the study with no changes in body weights, food or water consumption. The only observed clinical chemical change was an increase in serum cholesterol in the 1000× dose group. At the highest dose, the mixture exhibited mixed-type microsomal enzyme induction characteristics with increases in ethoxyresorufin *o*-deethylase, methoxyresorufin *o*-demethylase, pentoxyresorufin *o*-deethylase, benzyloxyresorufin *o*-dealkylation, aminopyrine demethylase and coumarin 7-hydroxylase activities. These changes were accompanied by increases in microsomal protein and percent liver weight per body weight.

A PBTK modeling framework for organochlorine mixtures has been developed, though we are still working to fit the toxaphene data in the model. The PBTK model was used to simulate the area under the concentration versus time curve (AUC) of parent chemical in rat liver, and this dose surrogate was related to the severity of hepatic damage observed during the experimental studies.

The PBTK model that simulates the AUC_{liver} that corresponds to NOAEL determined in the rat was scaled to a human. The exposure dose (i.e. TDI) that yields the same AUC_{liver} was determined.

The assessment of the potential risks to the Inuit populations exposed to these organochlorine compounds was performed using the population distribution data on the dietary intake of the chemicals in the mixture collected from the CINE dietary survey (Kuhnlein et al. 2000, this volume).

Results

Model structure

Physiologically based pharmacokinetic (PBTK) models have been developed and validated for several organohalogen substances in rodents and humans. These models describe the organism as a set of tissue compartments interconnected by systemic circulation and provide simulations of the tissue and blood kinetics of chemicals based on quantitative relationships among the mechanistic determinants (physiological, physicochemical and biochemical). The PBTK model used in the present project consisted of six compartments (adipose tissue, skin, richly perfused tissues, slowly perfused tissues, liver and blood) interconnected by arterial and venous blood pools. All tissue compartments were described as a composite of cellular matrix and blood sub-compartments such that diffusion limitation could be introduced as necessary. The dose to the animal was via oral administration, and clearance due to tissue uptake, macromolecular binding, metabolism and biliary excretion were accounted for.

Model parameterization

The rat body weight specified in the model corresponded to that of the animals used in the experimental study at Health Canada. Based on information on rat body weights, corresponding tissue volumes, cardiac output and tissue blood flow rates were calculated based on standard allometric relationships found in the literature (Table 1). The tissue:blood partition coefficients ($P_{t:b}$) for all chemicals in the mixture were calculated using the algorithm below (Poulin and Krishnan 1995).

$$P_{t:b} = \frac{(P_{ow} \cdot F_{nlt}) + F_{wt}}{(P_{ow} \cdot F_{nlb}) + F_{wb}} \qquad [1]$$

where P_{ow} = n-octanol:water partition coefficient, F = fractional volume, nl = neutral lipid, w = water, t = tissue, and b = blood. Since the log P_{ow} of chemicals investigated in the present study are higher than 3.3, Equation 1 simplifies to:

$$P_{t:b} = \frac{F_{nlt}}{F_{nlb}} \qquad [2]$$

The adequacy of the above equation for predicting $P_{t:b}$ of highly lipophilic organic chemicals has recently been verified (Haddad et al. 1999). Based on this equation, the $P_{t:b}$ of highly lipophilic organics (such as the ones investigated in the present study) is a constant, which is equal to the ratio of neutral lipids content of tissues and blood. Table 2 compares the $P_{t:b}$ predicted using the ratio of rat tissue and blood lipid levels with the values determined by fitting during previous PBTK modeling efforts. The hepatic extraction ratios (E) for the various chemicals were derived by running the PBTK model with E value set to zero initially, and then determining the value of E by iterative simulations and optimization of model fit to experimental data on blood concentrations measured at CINE (Table 3).

Table 1. Parameters used in the physiologically based toxicokinetic model in rat

Model Parameters	Symbols	Values
Body weight (g)	Bw	240
Cardiac output (mL·h^{-1})	Qc	4985
Tissue volumes (fraction of body weight)		
adipose	VFO	0.07
skin	VSO	0.16
liver	VLIO	0.04
slowly perfused tissues	VSPO	0.5
richly perfused tissues	VRPO	0.05
blood	VBO	0.09
Tissue blood volumes (fraction of tissue volumes)		
adipose	VFBO	0.05
skin	VSBO	0.01
liver	VLIBO	0.05
slowly perfused tissues	VSBO	0.05
richly perfused tissues	VRBO	0.01
Blood flow rate (fraction of cardiac output)		
adipose	QFF	0.07
skin	QSF	0.058
liver	QLIF	0.183
slowly perfused tissues	QSPF	0.4
richly perfused tissues	QRPF	0.289
Tissue diffusion coefficient (fraction of tissue blood flow rate)		
adipose	PAFF	1
skin	PASF	1
liver	PALIF	1
slowly perfused tissues	PASPF	1
richly perfused tissues	PARPF	1

Model simulations and validation

Simulations were conducted to simulate the dietary exposure experiment conducted at Health Canada. Accordingly the model simulated the exposure of rats to contaminant mixtures at three different dose levels (Table 4) in diet for 5 days per week for 4 weeks, and the sacrifice was made at 696 hours. The model simulations corresponded to the tissues and blood concentrations of the various chemicals in the mixture-fortified diet. The PBTK model was written as a program in Advanced Continuous Simulation Language® (Concord, Massachusetts), and solved using the parameter values specific to the rat and those that are specific to the chemicals in the mixture. The model simulations of the tissue concentrations

Table 2. Comparison of rat tissue:blood partition coefficients used in the unified model with those used in individual chemical models

Tissue	Tissue:Blood Partition Coefficients	
	Predicted*	Fitted**
Adipose	199	296.66 ± 130
Liver	12	12.66 ± 3.75
Skin	23	37.40 ± 16.68
Richly perfused tissues	12	11.61 ± 7.6
Slowly perfused tissues	3	3.52 ± 1.5

* Predictions were obtained by dividing the neutral lipid content of tissues with that of blood.

** The data represent mean ± standard deviation (n = 3) of partition coefficients obtained by fitting physiologically based toxicokinetic models.

resulting in animals exposed to various chemicals in the diet for a period of 4 weeks were compared with the experimental data measured at CINE. For conducting these simulations, no prior knowledge of the experimental data on tissue concentrations was necessary. Table 5 compares the a priori, independent predictions of the PBTK model with the corresponding experimental data on adipose tissue concentrations in the exposed animals.

Model application

The PBTK model was then applied to simulate the AUC and concentration of chemicals in the target organ, namely, liver of rats exposed to NOAEL determined in the study, i.e. the highest dose that did not induce any adverse effect (Table 6). Since the human data on the concentrations of chlordane, HCH (hexachlorocyclohexane), DDT and PCBs (polychlorinated biphenyls) have previously been reported for the family and not for individual isomers, the sum of AUC or concentrations for each chemical class was calculated. The sum of AUC for HCB (hexachlorobenzene), chlordane, HCH, DDT, mirex, PCBs and dieldrin corresponded to 391 974, 7701, 394 477, 1 913 572, 26 640, 1 502 456 and 235 516 μg·hr·L^{-1}, respectively. The value for toxaphene is not yet available. The concentration determined at the time of sacrifice in the liver of rats exposed to the dietary contaminants was observed to be close to steady-state. Therefore, the subsequent exercise involved the determination of the human exposure dose that corresponded to these liver steady-state concentrations associated with NOAEL in the rat (without the use of any additional uncertainty factors). For this effort, the rat model was scaled to a human model by substituting the model parameters with the species-specific values.

Table 3. Hepatic extraction ratios of chemicals in rats determined using physiologically based toxicokinetic models

Organochlorine Substances	Dose 1 E*	Dose 2 E	Dose 3 E
α-hexachlorocyclohexane	0.006	0.0023	0.0135
hexachlorobenzene	0.005	0.0022	0.0056
β-hexachlorocyclohexane	0.002	0.04	–
γ-hexachlorocyclohexane	0.013	0.0045	0.06
oxychlordane	0.0001	0.0001	0.0017
trans-chlordane	0.0001	0.016	0.1
cis-chlordane	0.004	0.03	0.18
trans-nonachlor	0.001	0.003	0.018
DDE	0.0001	0.0001	0.0049
dieldrin	0.0001	0.0048	0.018
cis-nonachlor	0.0175	0.022	0.045
DDT	0.0001	0.0001	0.0023
mirex	0.0001	0.0001	0.0001
PCB-28	0.004	0.004	0.028
PCB-32	0.045	0.04	0.135
PCB-99	0.012	0.0015	0.007
PCB-101	0.02	0.02	0.045
PCB-105	0.003	0.008	0.015
PCB-118	0.3	0.006	0.012
PCB-128	0	0.005	0.013
PCB-138	0.0008	0.0033	0.0095
PCB-153	0.015	0.007	0.013
PCB-156	0	0.003	0.015
PCB-180	0.0001	0.0045	0.0085
PCB-183	0.0001	0.011	0.014
PCB-187	0.0001	0.004	0.007
PCB-199	0.005	0.02	0.1

* Hepatic extraction ratios

Hepatic and blood concentrations in humans at steady-state corresponding to rat NOAEL and the equivalent reference dose in humans calculated with the PBTK model are presented in Table 7. The reference doses are compared to the current TDI used by Health Canada and the mean organochlorine intake levels estimated by the recent CINE dietary survey (Kuhnlein et al. 2000, this volume).

Table 4. Dose levels used in the Health Canada animal feeding experiments*

Organochlorine Substances	Dose 1 ($\mu g \cdot kg^{-1} \cdot day^{-1}$)	Dose 2 ($\mu g \cdot kg^{-1} \cdot day^{-1}$)	Dose 3 ($\mu g \cdot kg^{-1} \cdot day^{-1}$)
α-hexachlorocyclohexane	1.09	10.90	109.00
hexachlorobenzene	0.94	9.36	93.60
β-hexachlorocyclohexane	0.10	1.04	10.40
γ-hexachlorocyclohexane	0.01	0.96	9.60
oxychlordane	1.44	14.40	144.00
trans-chlordane	0.05	0.48	4.80
cis-chlordane	0.45	0.48	44.80
trans-nonachlor	1.09	10.92	109.20
DDE	3.39	33.90	339.00
dieldrin	0.92	9.20	92.00
cis-nonachlor	0.45	4.48	44.80
DDT	0.90	9.04	90.40
mirex	0.05	0.48	4.80
PCB-28	0.12	1.20	12.00
PCB-32	0.40	4.00	40.00
PCB-99	0.44	4.40	44.00
PCB-101	0.61	6.08	60.80
PCB-105	0.10	1.04	10.40
PCB-118	0.60	6.00	60.00
PCB-128	0.05	0.52	5.20
PCB-138	0.89	8.88	88.80
PCB-153	1.28	12.80	128.00
PCB-156	0.04	0.44	4.40
PCB-180	0.38	3.80	38.00
PCB-183	0.17	1.72	17.20
PCB-187	0.16	1.60	16.00
PCB-199	0.05	0.52	5.20

* Four groups of 12 female rats were administered the mixture in corn oil by gavage (three dose levels plus controls) for 28 days.

Discussions and Conclusions

This study is unique in that the chemical mixture used for the animal feeding experiment was prepared based on the concentrations of organochlorines found in an average Inuit diet as reported in Broughton Island in 1988 (Chan et al.1997). The risk assessment model developed is, therefore, relevant to the Inuit populations in northern Canada. A PBTK model was developed based on the results of the animal feeding experiment and the model was scaled to a human model. Reference doses of organochlorine were estimated by determining the human exposure dose that corresponded to these liver concentrations associated with NOAEL in the rat, i.e. the 1,000× organochlorine concentrations found in the mean Inuit diet. Unlike the classical paradigm used to establish the current TDI, this inter-species extrapolation was performed without the use of any additional uncertainty factors. In general, the exposure doses calculated using the human PBTK model were greater than the currently used acceptable daily intakes, except for DDT and chlordane, which were similar. Comparison of the more recent estimation of mean daily intake of organochlorines among the Inuit communities that participated in the CINE dietary survey to the reference doses estimated in this study suggest that the intake of chlordane remains 20 times higher than the calculated reference dose. This result suggests that the presence of chlordane in the Inuit diet may pose an increased risk to the people.

The model has now been adopted to simulate the pregnancy and lactation conditions, such that the simulations can in the future be compared with available data on cord blood and in children for risk assessment purposes.

Project Completion Date

September 2000

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Table 5. Comparison of experimental data with physiologically based toxicokinetic model simulations of adipose tissue concentrations in rats exposed to chemical mixture for 4 weeks

Organochlorine Substances	Dose 1 Fat Tissue Concentration		Dose 2 Fat Tissue Concentration		Dose 3 Fat Tissue Concentration	
	Mean ± SE* (ng·mL^{-1})	Sim**	Mean ± SE (ng·mL^{-1})	Sim	Mean ± SE (ng·mL^{-1})	Sim
α-hexachlorocyclohexane	403 ± 109	208	3586 ± 751	548	27 488 ± 4830	9228
hexachlorobenzene	597 ± 175	147	6745 ± 1983	1866	47 800 ± 8618	13 965
β-hexachlorocyclohexane	0 ± 0.0	21.1	0 ± 0.0	26.7	0 ± 0.0	–
γ-hexachlorocyclohexane	25 ± 16	7.5	73 ± 20	157	339 ± 59	1783
oxychlordane	726 ± 229	352	7505 ± 1410	3524	33 849 ± 8529	30 050
trans-chlordane	49 ± 15	4.96	85 ± 53	32.8	46 ± 11	29
cis-chlordane	21 ± 4	31.3	61 ± 34	47.4	106 ± 25	186
trans-nonachlor	239 ± 51	243	2731 ± 562	2028	8550 ± 1974	7106
DDE	2303 ± 571	829	17 643 ± 3749	829	95 664 ± 23 272	53 559
dieldrin	499 ± 173	225	2216 ± 448	1739	4888 ± 817	7106
cis-nonachlor	40 ± 8	30	422 ± 98	237	1716 ± 351	983
DDT	728 ± 159	220	4453 ± 1002	2212	20 972 ± 3686	17 867
mirex	14 ± 4	5.28	119 ± 25	117.5	551 ± 103	1174
PCB-28	19 ± 6	20.4	158 ± 38	204	1091 ± 169	482
PCB-32	5 ± 2	8.77	35 ± 10	102.8	226.4 ± 31	149
PCB-99	97 ± 29	41.1	930 ± 248	935	8830.8 ± 1679	5877
PCB-101	33 ± 7	35.5	334 ± 59	355	2236.1 ± 320	1334
PCB-105	20 ± 4	19.3	159 ± 32	128.7	1262.7 ± 196	804
PCB-118	124 ± 31	64.2	1278 ± 269	867	10 256.2 ± 1655	5605
PCB-128	96 ± 87	5.72	74.7 ± 17	75	546.5 ± 96	455
PCB-138	217 ± 53	201.4	2024 ± 422	1606	13 383.1 ± 2420	9850
PCB-153	236 ± 61	99	2514 ± 538	1715	19 945.4 ± 3631	11 242
PCB-156	8 ± 3	4.8	76 ± 17	81.7	596 ± 102	340
PCB-180	49 ± 16	92.1	558 ± 125	621	4878.3 ± 913	4532
PCB-183	18 ± 5	34.9	191 ± 42	172	1600.2 ± 287	1505
PCB-187	42 ± 13	39	259 ± 56	273	2256.7 ± 404	2137
PCB-199	4 ± 3	2.85	12 ± 3	30.4	77.6 ± 11	33.2

* Mean ± standard error (n = 6)

** Physiologically based toxicokinetic model simulations

Table 6. Comparaison of the predicted and experimental liver concentration and AUC of various chemicals in exposed animals

	Dose 3	Dose 3 Liver Tissue Concentration	Dose 3 Liver Tissue Concentration	Dose 3 Liver Tissue AUC 696 hours
Organochlorine Substances	(μg·kg^{-1}·day^{-1})	Experimental Mean ± SE* [μg·L^{-1}]	Sim** [μg·L^{-1}]	Sim [μg·h·L^{-1}]
α-hexachlorocyclohexane	108.8	1021 ± 103	567	326 650
hexachlorobenzene	93.6	2347.8 ± 283	831	391 974
β-hexachlorocyclohexane	10.40	0.0 ± 0.0	145	58 044
γ-hexachlorocyclohexane	9.6	8.7 ± 2	8.8	9783
oxychlordane	144.00	1399.7 ± 178	1740	732 801
trans-chlordane	4.8	1.0 ± 0.2	1.97	3059
cis-chlordane	12.8	1.9 ± 0.5	1.7	4642
trans-nonachlor	109.2	223.4 ± 64	442	279 546
DDE	339.2	2592.1 ± 430	3173	1 467 800
dieldrin	92.00	166.4 ± 33	372	235 516
cis-nonachlor	44.8	36.4 ± 13	62	58 865
DDT	90.40	811.4 ± 63	1040	445 772
mirex	4.80	111.3 ± 11	66	26 640
PCB-28	12	34.2 ± 5	30.2	22 859
PCB-32	40.00	4.0 ± 1.1	9.6	19 159
PCB-99	44.00	702.8 ± 77	353	172 730
PCB-101	60.80	81.4 ± 12	84	79 535
PCB-105	10.40	66.4 ± 5.1	49.7	29 544
PCB-118	60.00	542.5 ± 39	343	190 817
PCB-128	5.20	38.3 ± 12	23	14 056
PCB-138	88.80	994.7 ± 362	598	312 583
PCB-153	128.00	1452.3 ± 500	691	392 861
PCB-156	4.40	48.9 ± 15	21	12 499
PCB-180	38.00	428.8 ± 44	274	139 596
PCB-183	17.20	172.8 ± 17	86	50 093
PCB-187	16	184.7 ± 19	128	62 810
PCB-199	5.20	3.2 ± 0.4	2.1	3314

* Mean ± standard error (n = 6)

** Physiologically based toxicokinetic model simulations

Table 7. Hepatic and blood concentrations in humans at steady-state corresponding to rat no observable adverse effect level (NOAEL) and the equivalent reference dose in humans calculated with physiologically based toxicokinetic (PBTK) model*

Compounds	Steady-State Hepatic Concentration in Humans** ($\mu g \cdot L^{-1}$)	Steady-State Blood Concentration in Humans** ($\mu g \cdot L^{-1}$)	Reference Dose in Humans** ($\mu g \cdot kg^{-1} \cdot d^{-1}$)	Current TDI ($\mu g \cdot kg^{-1} \cdot d^{-1}$)	Mean Intake among Inuit communities ($\mu g \cdot kg^{-1} d^{-1}$)
Hexachlorobenzene	831	69.3	2.7	0.27	0.12
ΣChlordane	3.67	0.31	0.02	0.05	0.43
ΣHexachlorocyclohexane	721	60.1	4.4	0.3	0.03
ΣDDT	4213	351	18	20	0.53
Mirex	66	5.5	0.21	0.07	0.01
ΣPCBs	2692.9	224.4	17	1	0.66
Dieldrin	372	31	18.5	0.1	–

* The reference doses are compared to the current tolerable daily intake used by Health Canada and the mean organochlorine intake levels estimated by the recent CINE dietary survey.

** Determined with PBTK model.

Effects of Prenatal Exposure to Organochlorines and Mercury on the Immune System of Inuit Infants

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Abstract

This project, nested in the ongoing study *PCBs and Infant Development*, examines the effects of prenatal and postnatal (from birth to 12 months of age) exposure to organochlorines and mercury on the immune system of Inuit infants. To date, 56 Inuit mothers and their newborns from Nunavik are participating in the study and another 80 should be enrolled over the entire study period. Discussion with Greenlandic/Danish researchers is under way and, if an agreement is reached, it will be possible to include 70 Greenlandic infants to reach a total sample size of 150 participants. Immune parameters have not been analyzed so far in blood samples since analyses are usually done when a large number of samples have been collected. Nevertheless, some samples have been analyzed for vitamin A and preliminary results show median vitamin A concentrations in blood samples of 25 mothers, 10 infants and 11 cords of 41.4 $\mu g \cdot dL^{-1}$, 28.1 $\mu g \cdot dL^{-1}$ and 16.6 $\mu g \cdot dL^{-1}$, respectively. The collection of perinatal information and a look at the general health status of the children during the first year of life, which includes the incidence of infectious diseases obtained from medical charts, will start next fall.

Objectives

Long-term

- To examine the effects of prenatal and postnatal exposure to organochlorines (OCs) and mercury (Hg) on the immune system of Inuit infants, from birth to 12 months of age.

Short-term

1. To assess prenatal and postnatal exposure to OCs and Hg.
2. To measure immunological parameters: humoral immunity, cellular immunity
3. To assess vitamin A status in Inuit infants

Introduction

Substantial information concerning the contamination of northern marine food by persistent organic pollutants and heavy metals is now available (Barrie et al. 1992, Braune et al. 1999, Bright et al. 1995, Burkow and Kallenborn 2000, Lockhart et al. 1992, Muir et al. 1999, Muir et al. 1992, Thomas et al. 1992). Chemicals such as pesticides (dieldrin, mirex, toxaphene), industrial compounds and by-products of industrial processes (HCB [hexachlorobenzene], PCBs [polychlorinated biphenyls], PCDDs [polychlorinated dibenzo-*p*-dioxins], PCDFs [polychlorinated dibenzofurans], as well as heavy metals (inorganic and organic Hg, lead, cadmium) have been emitted throughout the world for many years. Since the late 1970s many regulatory actions have been taken to limit their emission but they are still being released due to improper storage and ongoing use in certain parts of the world.

Prenatal and postnatal exposures to organochlorines (OCs), in particular dioxin-like compounds, produce a wide variety of toxic effects on animals and humans. Alterations of T-cell subsets, serum immunoglobulin A (IgA) and immunoglobulin M (IgM) concentrations, delayed-type hypersensibility (DTH), and complement activation have all been documented in rodents, primates and humans (Chang et al. 1982, Esser and Welzel 1993, Hoffman et al. 1986, Holsapple et al. 1984, Lu and Wu 1985, Neubert et al. 1992, Thomas and Hinsdill 1979, Tryphonas et al. 1991a, Tryphonas et al. 1991b, White et al. 1986). The development of the immune system in utero and during infancy is particularly sensitive to immunotoxic products such as OCs. High exposure during early life could lead to permanent defects of the immune system and thus decrease resistance to infectious agents (Badesha et al. 1995).

Organic and inorganic Hg may induce cytotoxic activity in cellular components of the immune system in several species of rodents. Methylmercury, a form of organic mercury, can alter non-specific defense mechanisms, such as inhibition of natural killer cell activity in rats and mice. It also decreases the expression of certain activation markers of T cells (HLA-Dr, IL-2R) (Moszczynski 1997). Moreover, it has been well demonstrated that methylmercury can affect the functions of B cells and therefore reduce the humoral mediated response (Moszczynski 1997). Exposure to inorganic mercury induces allergies and autoimmune problems in hypersensitive individuals (Bagenstose et al. 1999).

The high incidence of infectious diseases (in particular, meningitis, broncho-pulmonary and middle ear infections) in Native infants and children from Nunavik has been known for many years (Dufour 1988, Proulx 1988). The middle ear infections are of special concern since approximately 25% of school-age Inuit children suffer some kind of hearing loss (Julien et al. 1987). Because of the immunotoxic effects of OCs and methylmercury, particularly after perinatal exposure, we hypothesized that part of the high infection incidence among Inuit infants could be related to the high maternal body burden of these contaminants.

This study is nested in the ongoing National Institute of Environmental Health Sciences neurodevelopmental cohort study, and will involve approximately 150 Inuit mothers and their newborns. It investigates possible detrimental effects on the immune system of Inuit infants that may be induced by prenatal and postnatal breast-feeding exposure to OCs and Hg. Immune system function is evaluated using several immunological markers including the level of antibody produced by the infant following *Haemophilus influenza* immunization, the level and function of the complement system and its components, and the level of some Th1 and Th2 cytokines. Vitamin A concentration is also evaluated in the mother and the infant since striking similarities exist between symptoms of high dioxin-like compound exposure and vitamin A deficiency. Finally, infectious episodes are recorded over the first 12 months of life and possible relationship between recurrent infections and high OC burdens will be analyzed. Results of this research could be used by intervening institutions for the purpose of risk prevention and management, by other researchers, and by the public.

Activities in 1999-2000

The training of the fieldwork coordinators based in Puvirnituq and Inukjuak was completed. With the collaboration of a nurse and a laboratory technician based in Puvirnituq, some laboratory procedures were explained and rehearsed by the new field coordinators in June 1999 and March 2000.

In December 1999, a new fieldwork coordinator for Inukjuak was hired and trained (maternity leave replacement). This person is responsible for participating in the recruitment of pregnant women, for ensuring that blood samples are collected (maternal blood samples during labour and infant blood samples at 6 months old), and for following laboratory procedures for the conservation of specimens before analysis.

To date, 66 pregnant women have been recruited and 56 mothers and their newborns have been enrolled in the study; 56 maternal and 34 infant blood samples have been collected.

In order to reinforce the links between the Inuit communities and the health professionals who are involved in the study, the project coordinator (a research nurse based in Quebec City) meets once a year in every village with community members, midwives, nurses and all medical professionals who are involved in the data collection. These meetings stimulate interesting discussions and give the research nurse the opportunity to answer questions after informing them about the project, ongoing activities, difficulties met and the support needed. During the last trip, in November 1999, the project coordinator participated in a local FM-radio phone-in show broadcast to let the communities know more about the project and to answer some of their questions. The researcher-coordinator of the Infant Development Study, the Nunavik Nutrition and Health Committee coordinator, and one researcher also participated in this activity.

Data collection is progressing but the recruitment of new participants and the number of specimens collected decreased last fall, mainly because of the absence for many months of a field coordinator. Consequently, to date, the number of participating pregnant women is 66. We will also evaluate the anti-Hib response of 27 additional infants who were already enrolled in the ongoing neuro-developmental cohort study prior to the initiation of the immune function study, using stored blood samples collected at 6 months of age. Hence, it will be possible to evaluate the antibody response against *Haemophilus influenza* type b (Hib) for approximately 100 Inuit infants from Nunavik.

Results

Immune parameters have not yet been analyzed in blood samples since the current sample size is not large enough. These analyses will be performed in one batch to avoid between-days variations.

Vitamin A analysis was performed on 25 maternal blood, 10 infant blood and 11 cord blood samples. The median vitamin A concentration was 41.4 $\mu g \cdot dL^{-1}$, 28.1 $\mu g \cdot dL^{-1}$ and 16.6 $\mu g \cdot dL^{-1}$ for maternal, infant and cord blood samples, respectively. Although the sample size is small, results indicate that 4% of the mothers and 10% of the infants are within the 10 to 20 $\mu g \cdot dL^{-1}$ range indicating mild vitamin A deficiency.

Discussion and Conclusions

Although the sample size is small, results indicate that 4% of the mothers and 10% of the infants are within the 10 to 20 $\mu g \cdot dL^{-1}$ range indicating mild vitamin A deficiency. Vitamin A concentration in liver has been shown to decrease in relation to PCB body burden in wildlife species. The sample size in the present study was too small to investigate the possible correlation between vitamin A plasma levels and PCBs.

In order to meet the objectives of this study, we will need a cohort of approximately 150 mothers and infants. Recruitment began in February 1999. With the experience of this year, because the number of births per calendar year is limited and that only two communities of Nunavik participate in this immunological study, we do not expect to have more than 80 mothers and infants enrolled over the entire study period: four in 1998-1999, 56 in 1999-2000 and 12 in 2000-2001 in Nunavik. The recruitment of pregnant women should end in fall 2000. Collection of samples (6 months of age) will end in fall of 2001. The project should be completed in March 2002. Consequently, the creation of an extension of this study in Greenland is needed to ensure that our objectives will be met. Discussion with Greenlandic/Danish researchers is under way to include 70 Greenlandic infants, allowing to reach a total sample size of 150 participants. Since we need to have comparable data, all analysis will be performed in our laboratory.

Expected Project Completion Date

March 2002

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Mercury in Salluit: Temporal Trend and Interaction with Selenium

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Abstract

At the request of the Municipal Council of Salluit (Hudson Strait, Nunavik), exposure of Sallumiut to mercury was assessed through analyses of hair and blood samples during 1998-1999, some 20 years after surveys conducted by Health Canada had indicated high blood mercury levels in this population. Close inspection of the data revealed that mercury exposure in 1978 was exceptionally high, probably due to the large beluga harvest of 1977. Similar mercury concentrations were documented during subsequent surveys (1979, 1980 and 1999). In 1999-2000, markers of oxidant stress were measured in blood samples (glutathione peroxidase and glutathione reductase) and exhaled breath samples (pentane concentration). Pentane levels in exhaled breath could not be interpreted due to the large between-series variability in exhaled breath and ambient air concentrations. Blood mercury concentrations were inversely correlated to glutathione reductase activities ($r = -0.23$, $p = 0.005$) while plasma selenium levels were positively correlated to glutathione peroxidase activities ($r = 0.16$, $p = 0.05$). Hence, both mercury and selenium appear to modulate the activity of enzymes involved in oxidant stress response.

Objectives

1. To reassess mercury (Hg) exposure in 150 Sallumiut who participated in the biological monitoring survey of Health Canada during 1978-1980, using hair and blood samples.
2. To assess selenium (Se) status in participants using hair, blood and urine samples.
3. To assess antioxidant status and oxidative stress biomarkers in blood and exhaled breath of Sallumiut.

Introduction

During a series of surveys conducted by Health Canada between 1977 and 1982, analysis of samples from 142 Nunavik residents revealed a mean blood Hg concentration of 240 nmol·L^{-1} (range = 21 to 1269 nmol·L^{-1}) (Wheatley and Paradis 1996). In general populations consuming little or no fish or marine mammals, blood Hg values are generally around 10 nmol·L^{-1}. For example, a recent survey revealed that blood concentrations in 1127 healthy American men averaged 10 nmol·L^{-1} (Kingman et al. 1998). Residents of Salluit were found to have particularly high levels with 17% of participants showing concentrations > 500 nmol·L^{-1}, whereas in other Nunavik communities only 2% of concentrations exceeded that level. At the request of the Municipal Council of Salluit, the present study was set up to verify contemporary levels of Hg in adults who were tested at the end of the 1970s.

Hg is an environmental contaminant originating from both anthropogenic and natural sources. It can be transported from distant sources to the Arctic by atmospheric and oceanic transport. Hg has been found in all components of the Arctic ecosystem and is mainly present as methylmercury (MeHg) in fish and marine mammals (Muir et al. 1992). Consumption of fish and marine mammals constitutes an important source of exposure, especially in populations such as the Inuit that depend on marine species for sustenance.

MeHg is a highly toxic environmental neurotoxin that can cause irreversible damage to the central nervous system, and to which the developing fetal brain is especially sensitive (Choi 1989, Clarkson 1993, Clarkson 1997). The lowest dose of MeHg that may impair neurodevelopment in the human fetus is not known. The chief concern is with effects arising from prenatal exposure to MeHg, such as early sensory motor dysfunction (e.g. delayed onset of walking and talking) and delayed development and cognitive changes in children. Although the underlying biochemical and molecular mechanisms that lead to impaired nerve cell function and nerve cell degeneration are not well understood, there is abundant evidence supporting the hypothesis that a major mechanism of MeHg neurotoxicity involves an oxidative stress (Sarafian et al. 1991, Yee et al. 1996). Mercurials promote increased production of reactive oxygen species (ROS) via deregulation of mitochondrial electron transport as well as through glutathione (GSH) depletion (Lund et al. 1993). The oxidative stress hypothesis is clearly supported by the finding that MeHg neurotoxicity can be inhibited by various antioxidants including Se (Park et al. 1996) and N-acetyl-L-cysteine a precursor of GSH (Ornaghi et al. 1993).

Se has been shown to counteract the toxicity of MeHg in many experimental systems (reviewed in Whanger 1992) but the mechanism(s) involved is (are) poorly understood. On the one hand, Se known to bind directly to Hg and thus participate in its sequestration. On the other hand, Se supplementation is also known to boost the cellular antioxidant defence by stimulating the synthesis of several Se-dependent enzymes including cytoplasmic glutathione peroxidase (GSHPx), a key enzyme in the detoxification of peroxides and prevention of oxidative DNA damage (Mirault et al. 1991, Mirault et al. 1992). In the second phase of this study, GSHPx activity was determined in blood samples of participants. We also measured the activity of glutathione reductase (GSHRd), another key enzyme involved in the defence against ROS, which is involved in regenerating GSH from glutathione disulfide. In addition, the level of pentane in exhaled breath was measured as an index of lipid peroxidation (Kneepkens et al. 1994). Lipid peroxidation is induced by MeHg administration in experimental laboratory animals (Huang et al. 1996). Hg-induced lipid peroxidation may be responsible, in part, for the high death rate from cardiovascular diseases observed in Eastern Finland (Salonen et al. 1995).

Activites in 1999-2000

Glutathione peroxidase and glutathione reductase activities

Activities of GSHRd and GSHPx were determined in plasma samples using commercial kits from Randox Laboratories Ltd. and an RA-XT automated spectrophotometer, in the CHUL biochemistry laboratory. In the GSHPx assay, GSHPx catalyses the oxidation of GSH by cumene hydroperoxide, in the presence of GSHRd and NADPH. Glutathione disulfide (GSSG) is immediately reduced to GSH, by concomitant oxidation of NADPH to NADP$^+$. In the GSHRd assay, GSHRd catalyses the reduction of GSSG in the presence of NADPH, which is oxidized to NADP$^+$. In both assays, enzyme activity is measured by following the rate of decrease in NADPH absorbance at 340 nm, and expressed in U·L^{-1} (1 unit = 1 μmole NADPH oxidized·min^{-1} at 37°C).

Pentane analysis in exhaled breath

Participants were required to breathe filtered air through a mouth piece while wearing a nose clip, in order to wash out ambient air hydrocarbons from the lungs. Exhaled breath was then collected in a Tedlar bag (20 L) using a unidirectional valve (Hans Rudolph). The air was then transferred from the bag to a glass vial for storage and transport. Pentane was measured directly in the glass vial using a "Headspace" sampler coupled to a gas chromatograph equipped with a capillary column (RTX-624) and a flame ionisation detector.

Temporal trend of mercury exposure

At the end of 1999, contacts were made with the Community Health Representative of Salluit and with the research assistant involved in the 1998-1999 data collection. With their help, different communication activities were organized. First, a personal letter was addressed to all participants to inform them of their results and provide appropriate recommendations. Included with this letter was a graph that allowed all participants to compare their 1999 hair Hg levels with that documented in previous Health Canada surveys (1978-1980). Second, a fact sheet pertaining to Hg contamination of the food chain addressed the following questions: "What is mercury? How does mercury get in your body? Are there any health risks linked to mercury contamination? Should we be concerned about the levels found in Sallumiut? What should we do?"

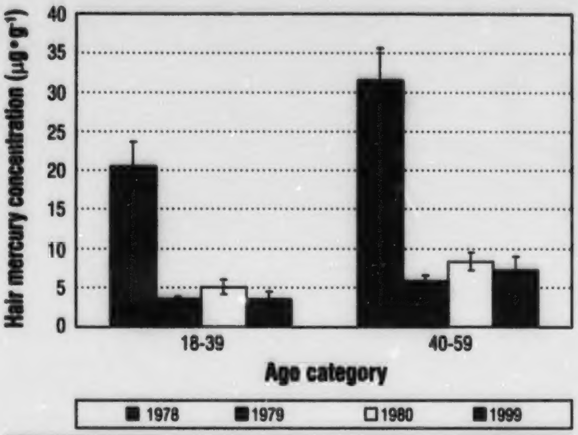
In March 2000, the principal investigator, another project team member and a Health Canada representative went to Salluit to participate in different communication activities. First, the study results were presented to the nursing personnel. We made sure that if participants called to discuss their results with a nurse, the same information would be given by all nurses. Second, the research team presented the results to the Municipal Council and Local Health Committee members. Finally, a phone-in show was organized so that the community could share their concerns and questions with the research team.

Results

Hair mercury analysis

Initial results of Hg analysis in hair samples, as reported in Dewailly et al. (1999), suggested that hair Hg levels were substantially lower in 1999 than at the end of the 1970s. The mean (geometric) Hg concentration in first-centimetre hair samples collected in 1999 was 6.1 $\mu g \cdot g^{-1}$ (95% confidence interval (CI) = 5.2 to 7.1 $\mu g \cdot g^{-1}$), a value 2.5-fold lower ($p < 0.001$) than that of 15.2 $\mu g \cdot g^{-1}$ (95% CI = 13.2 to 17.5 $\mu g \cdot g^{-1}$) determined in a series of surveys conducted by Health Canada surveys between 1978 and 1980. However, further analysis of the data revealed that only levels measured in 1978 were higher than those measured in 1999 (Figure 1). Mean (geometric) Hg concentrations measured during 1999 in the 18- to 39 year-old and 40- to 59-year-old-age groups were five and four times lower ($p < 0.001$), respectively, than those documented during the 1978 survey. The beluga harvest in Salluit was particularly large in 1977, when 104 animals were killed. In 1978, 1979 and 1980, numbers of beluga harvested were 36, 42 and 50, respectively. The year 1978 was also exceptional for the harvest of other species such as lake trout and seal. Higher consumption related to large harvests could explain why high Hg concentrations were found in hair samples in the spring of 1978.

Figure 1. Hair mercury concentrations (first cm) in Sallumiut, 1978-1999



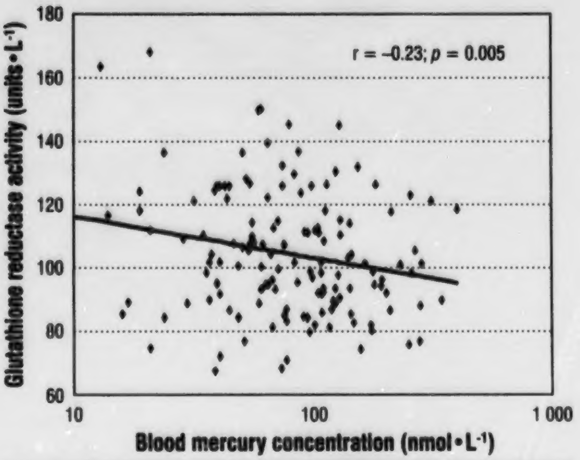
Markers of oxidative stress

GSHPx and GSHRd activities were measured in blood samples from 142 Sallumiut. Unfortunately, delays during shipment of samples from Salluit to our laboratory caused blood samples to thaw and led to significant losses of enzyme activity. Mean loss of activity was estimated for each enzyme by reproducing in the laboratory the storage conditions that prevailed during shipment (GSHPx: –32%; GSHRd: –9%), however, the inter-individual variability in activity loss is likely to have increased significantly the random error of the data.

Mean GSHRd activity in blood samples of the 142 participants was 104.8 $U \cdot L^{-1}$ (95% CI = 101.5 to 108.1 $U \cdot L^{-1}$). Figure 2 shows the statistically significant negative correlation between Hg blood levels (log-transformed) and GSHRd activities ($r = -0.23$, $p = 0.005$). Mean GSHPx activity in blood samples of participants was 1629 $U \cdot L^{-1}$ (95% CI = 1577 to 1682 $U \cdot L^{-1}$). A marginally significant positive correlation was noted between plasma Se concentrations and GSHPx activities ($r = 0.16$, $p = 0.05$). GSHPx activities were correlated to GSHRd activities ($r = 0.27$, $p = 0.001$).

Figure 3 displays concentrations of pentane measured in exhaled breath samples of participants and in ambient air samples, according to sampling date. The first series of breath samples contained low pentane levels (in most

Figure 2. Correlation between blood mercury concentration and glutathione reductase activity

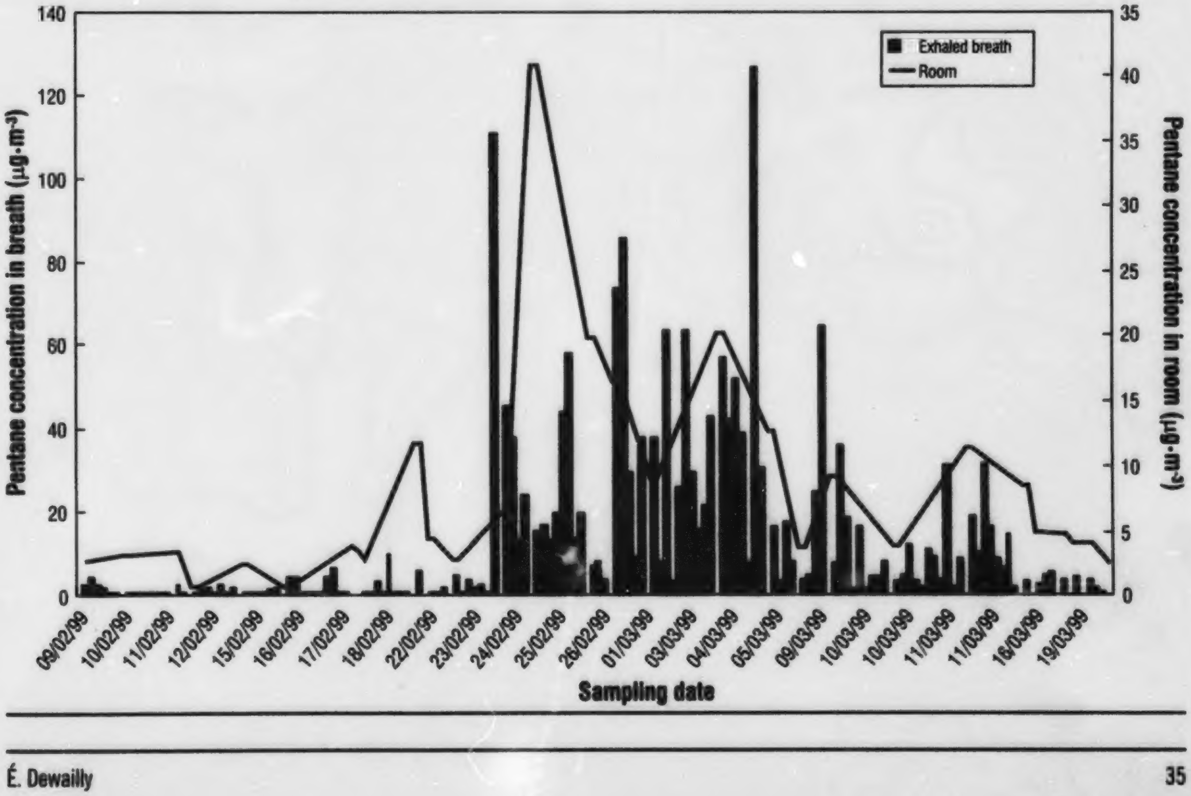


cases < 1.4 μg·m^{-3} detection limit of the analytical method) and room air contained in general < 5 μg·m^{-3}. However, results from the second series of breath samples (starting on February 24, 1999) were much higher and, in several cases, reached levels > 50 μg·m^{-3}. In parallel, concentrations measured in the room also rose to reach 40 μg·m^{-3} on February 25 and slowly declined thereafter. Considering the striking relationship between ambient air and exhaled breath pentane levels, results could not be interpreted in relation to Hg and Se concentrations.

Discussion and Conclusions

Results from the additional data analysis performed this year indicate that Hg exposure of Salluit residents is still high compared to southern populations, but is not different from that documented in 1979 and 1980. Hg exposure encountered in 1978 was exceptionally high, probably due to an unusually large harvest of beluga and other highly contaminated species in 1977. It has been suggested that the high concentrations of Se found in blood samples (Dewailly et al. 1999) may protect the Inuit population against Hg toxicity. Activities of enzymes involved in detoxication of free radicals were measured in order to investigate relations between Hg, Se and oxidative stress. We observed that Hg was negatively correlated to GSHRd activity, an NADPH-dependent enzyme that regenerates GSH from GSSG. In contrast, plasma Se concentration was positively correlated to GSHPx activity, a selenoenzyme that catalyses the conversion of hydrogen peroxide to water. Hence, Hg exposure may diminish defence mechanisms against oxidative stress by limiting the availability of GSH, while Se may afford protection by favouring the destruction of hydrogen peroxide.

Figure 3. Ambient air and exhaled breath pentane concentrations according to the date of sampling



Expected Project Completion Date

This project was completed on March 31, 2000. A second project is underway to investigate possible alterations in sensorimotor function in Sallumiut and in Hydro-Quebec workers seasonally exposed to Hg through sport fishing.

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Decision Making and Diet in the North: Balancing the Physical, Economic and Social Components

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Abstract

In the first year of this 2-year project, a collection, review and summary of past research done in northern regions relating to the social, cultural and spiritual benefits of traditional diets and diet-related activities (hunting, collecting, sharing, preparing, consuming) was conducted. Qualitative content analysis was used to identify common themes, to order and organize this information, and to identify common benefits related to different traditional/country foods and traditional/country food-related activities. Simultaneously, a culturally specific, community-based questionnaire was developed and tested in one region (Nunavik) to provide a quantitative assessment of the determinants of food choices. Use of this survey tool will help to understand the dynamics underlying changes in consumption of traditional/country food, and put in perspective the impacts of communication activities related to contaminants research. The framework and survey tool being developed in this project will be applicable to other northern regions and groups to better support health decision making, protection and promotion activities.

Objectives

1. To review and organize the reported social and cultural benefits and risks associated with a traditional diet and related activities (hunting, distribution, preparation, consumption).
2. To develop and apply a survey tool to increase our understanding of the determinants of diet behaviour.
3. To develop a conceptual framework for the ordered presentation of this information.
4. To link this framework with those organizing information on health and economic benefits and risks associated with traditional/country foods.

Introduction

While Western science has recently recognized the biological and economic benefits and risks related to traditional/country foods, traditional knowledge has long since recognized the many benefits of a traditional diet in terms of physical, social and spiritual well-being (Borré 1991, Borré 1994, Egede 1995). Others (Santé Québec 1992, Wheatley and Wheatley 1981, Wenzel 1986) have reported on the loss or disruption of these social and cultural aspects because of confusion and concern over food safety resulting from reports of environmental contamination. Northern health professionals, government representatives and advisory bodies have the responsibility to "manage," protect and promote both public health and the relationship between northern people and local resources, through balancing available information, providing advice and, in some cases, carrying out intervention programs. This management process has proven to be a difficult and, at times, challenging exercise, in particular because the needs, expectations and personal preferences (food, economic, cultural, etc.) of the population have not always been considered.

Aspects of physical and socio-economic health relating to traditional/country food sources in the North have been presented in organized frameworks which support the decision-making activities of health promoters and protectors and others with advisory responsibilities on environmental health issues in the Canadian Arctic (Dewailly et al. 1996, Dewailly et al. 1998). However, no work to date has focussed on presenting the social and cultural aspects related to traditional/country foods in a similar way. Similarly, little work to date has focussed on gaining a better understanding of the determinants of food choice in the North.

It is recognized that the social components cannot be compared, contrasted and weighted in the same quantitative manner as those of a physical health and economic nature. However, it is argued here that an organization and presentation of this information would be of value to northern managers and health professionals. Such an organization would allow for an explicit and ordered consideration of the reported social, cultural and spiritual benefits that Inuit associate with traditional/country foods and food-related activities, for use in the development of interventions and promotions to support nutritional health and well-being. The balanced consideration of all components of a traditional diet is vitally important to any decision regarding traditional food protection and promotion. These decisions directly impact individual health and well-being, and determine the extent to which the population is able and willing to comply to health policies, programs and interventions.

Those responsible for the protection and promotion of public health in the North must consider and have access to all available information in order to make informed decisions. This is becoming increasingly more important as the political, environmental health and economic issues surrounding traditional/country foods in the North are becoming increasingly complex (e.g. as we identify and learn more about new contaminants and their potential impacts on human health, as northern peoples strive for self-determination and subsequent organizational jurisdictions and mandates dealing with environmental health issues evolve, etc.).

Activities in 1999-2000

Part A: Review and organization of the reported social and cultural benefits and risks

Documents were located from electronic databases, specialized bibliographies, indexes to special journals and by manual library searches. The literature included academic articles, government reports, occasional publications and "grey" literature from organizations. The focus of the documents retained was Canadian Inuit, however, reports from other regions were included when the information was directly related to the subject of social, cultural or spiritual benefits of traditional/country foods. Documents were then scanned for passages containing reference to the subject of the study and entered into an electronic database. Categories were developed based on references made in passages to a particular activity (e.g. collection), its significance (e.g. cultural), the theme of that significance (e.g. identity), and its effect (e.g. definition) mentioned (Table 1). These groupings were developed and adapted, evolving as the review progressed (Miles and Huberman 1984, Tesch 1990). A numerical representation of the thematic breakdown of the discourse is presented in the results to show relative emphasis in the literature on certain areas; no statistical significance or importance of one category over another is implied.

Part B: Development of the survey tool

The development of the survey tool to identify and assess determinants of diet behaviour was undertaken through the following steps.

First, literature was reviewed and interviews were prepared using data collected in Kuujjuaq, Nunavik. A total of 18 individual interviews were conducted, comprised of four health representatives, three government representatives, one regional authority, nine food suppliers and one education representative.

Table 1. Categories developed and used in cataloguing citations

Activity	Significance	Theme	Effect
Collection	Cultural	Community	Continuity and expression
Consumption	Social	External influence	Definition
Distribution	Spiritual	Heritage	Increase
Preparation		Identity	Maintenance
		Individual	
		Well-being	

Second, seven group discussions (n = 33) were held with residents representing three age groups among men (13 to 44 years old) and four age groups among women (13 to 84 years old). Discussions focussed on the basic factors influencing their daily food choices, such as what was preferred and what was available.

Third, content analysis of the data from the discussion groups was conducted (Gagné and Godin 1999).

Fourth, the first draft of the questionnaire was produced. The draft was discussed with and reviewed by the Nunavik Nutrition and Health Committee and the northern research partner.

Fifth, the questionnaire was translated from English to Inuktitut and back-translated from Inuktitut to English by two different translators. The two versions were reviewed and the questionnaire was adjusted as required.

Next, the questionnaire was tested and re-tested with representatives from different groups of the population. A total of 26 women (ages 14 to 66) and 13 men (ages 14 to 79) participated in the test and re-test process. Nineteen individuals completed the English version while 20 completed the Inuktitut version of the survey during the first test stage. Nineteen and 20 individuals, respectively, completed the English and Inuktitut versions of the survey again in the re-test phase. Verification of the internal consistency of the theoretical variables (construct validity) was conducted after the first test phase using Cronbach's Alpha test (Cronbach 1951). Test of the stability of the instrument over time (reliability) was done by conducting an intra-class correlation analysis on the results from test 1 versus test 2 (Shroutte and Fleiss 1979).

Lastly, final versions of the questionnaires were produced (one English, one Inuktitut).

Results

Part A: Review and organization of the reported social and cultural benefits and risks

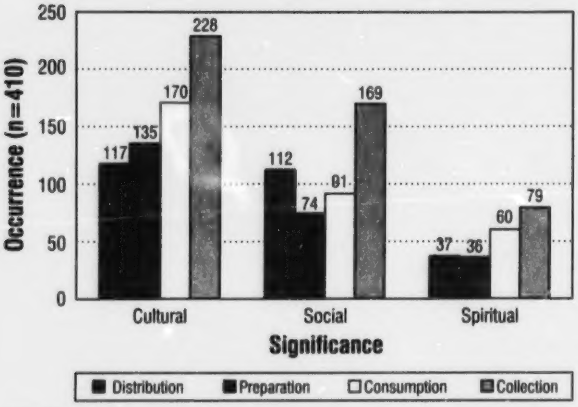
Four hundred and ten citations identified from the 112 sources were retained. Publication dates of articles ranged between 1967 and 1998, with a mean date of 1993. The last 30 years were chosen as the focus of this study in order to understand the topic from a more contemporary perspective. Not all citations were mutually exclusive in their reference to one category or another, as some were placed in more than one category or theme; however, many were clearly identified and defined by the categories developed in this review. The majority of citations were related to collection activities of traditional/country foods, their cultural significance, the heritage or traditional aspects of this significance, and the maintaining effects they have on this significance (Table 2).

The activities related to traditional/country foods are most often mentioned in reference to their cultural significance (Figure 1). Within the social, spiritual and cultural significance categories identified, a number of themes are commonly mentioned (Figure 2). Many of the citations making reference to the cultural significance of country foods, did so with regards to their value in terms

Table 2. Percent of total citations in discourse identified by category and sub-category

Categories	Sub-category	Percent of total citations / category
Activity	Collection	38
	Distribution	20
	Preparation	17
	Consumption	25
Significance	Cultural	49
	Social	35
	Spiritual	16
Theme	Community	22
	External Influence	2
	Heritage	32
	Identity	15
	Individual	10
	Well-being	19
Effect	Maintenance	53
	Continuity and Expression	16
	Definition	26
	Increase	5

Figure 1. Significance of traditional/country food related activities as reported in the literature



of heritage, identity, community or well-being. Many of those making reference to the social benefits of country foods did so in reference to their importance to community (social interactions), heritage or well-being, while spiritual importance was often reported with regards to its aspects of heritage (Figure 2). These foods and their specific related activities are reported to define and maintain aspects of social, cultural and spiritual life for Northerners (Figures 3 and 4). Maintenance and definition of these aspects was mentioned more often in relation to all country foods than was their provision of continuity or increasing these aspects in northern communities and lives (Figure 4).

Figure 2. Themes within reported significance of traditional/country foods

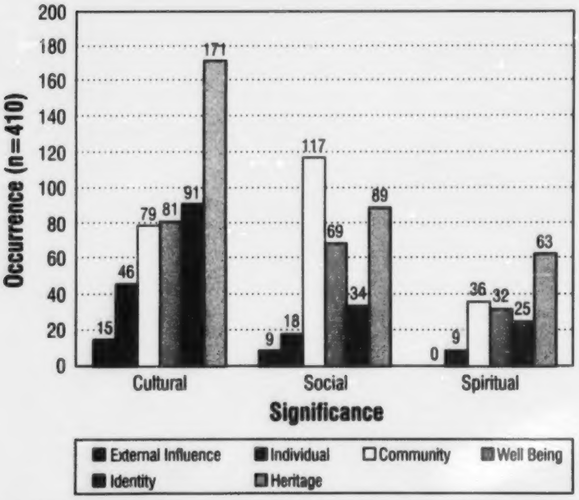


Figure 3. Effects on significance of traditional/country foods as reported in the literature

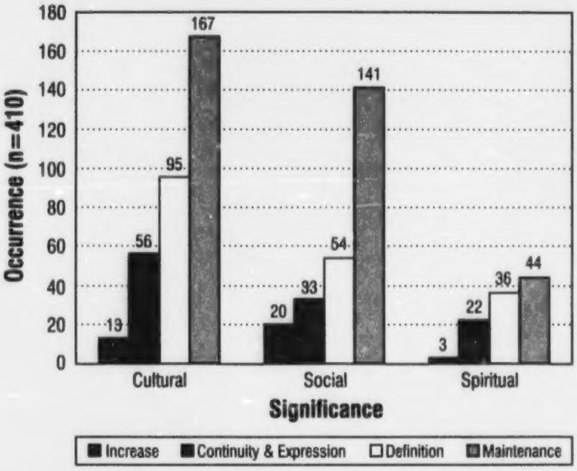
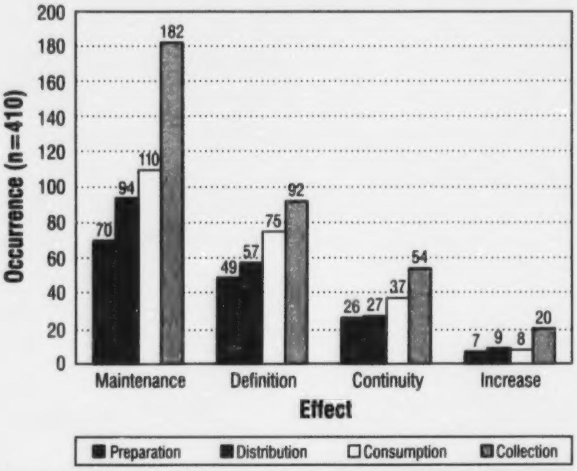


Figure 4. Effects of specific traditional/country foods related activities as reported in the literature



Part B: Development of the survey tool

The questionnaire developed was based on the emic content (as elicited from the population through group interviews) of the etically defined constructs of a model of behaviour prediction: Triandis' theory of interpersonal behaviour (1980) (Table 3).

Table 3. Categorized content of focus group discussions on which questionnaire was developed

	Potential determinants of decreased use	Potential determinants of increased use	Uncertain effects
Country food	*Affect/feeling:* • Tired of it too often/need for change *Beliefs:* • Fear of food poisoning (by *igunaq*) *Barriers/facilitating factors:* • Unavailability of resources • Scarcity of animals • Distance from community • Season • Community freezer empty • Money (expensive to buy, to harvest) • Lack of knowledge/ food preparation	*Affect/feeling:* • Taste • Cravings *Beliefs:* • Health/nutrition benefits • Strength/energy • Makes me feel *Inuk* • Fulfilment *Barriers/facilitating factors:* • Preparation (adding condiments, new recipes) • Cheap/free • Freshness • Tradition (being brought up with it, habit) • Sharing	*Normative beliefs:* • Elders • Health professionals • Parents *Role beliefs (reference groups):* • Older people • People with no income • Youth • People who hunt/fish • Inuit
Store-bought food	*Affect/feeling:* • Tired of one kind too often *Beliefs:* • Junk food unhealthy Less fulfilment *Barriers/facilitating factors:* • Cost (expensive) • Lack of freshness	*Affect/feeling:* • Taste/preference • Cravings *Beliefs:* • Health/nutrition benefits *Barriers/facilitating factors:* • Cook free /fast • Availability/always there • Variety/choices	*Normative beliefs:* • Health professionals • Spouse/children • Parents • TV commercials *Role beliefs (reference groups):* • Youth • People with income

Discussion and Conclusions

Part A: Review and organization of the reported social and cultural benefits and risks

Significant benefits are attributed to all traditional/country foods, with the literature being more comprehensive and descriptive in regards to the social and cultural components. Similarly, distinct benefits are attributed to the activities of collection, distribution, preparation and consumption. The literature reflects the differences between these activities. Each have their own respective importance associated with them as reflected in the literature. Despite the rich nature of the literature on these topics, it appears that relatively little attention is given to studies directly reporting the specific aspects of social or cultural importance attributed to these foods. Many studies simply report that social, cultural or spiritual significance exists while providing little empirical support for their argument. However, a specific treatment of this issue is complex as the nature of the benefits attributed to traditional/country foods is not easily identified, described, or in some way measured, as reflected in the literature reviewed here. Still, a description of this nature is helpful to understand the specific benefits attributed to these sources of health and well-being in contemporary Aboriginal communities and supports their explicit consideration in decision-making events on traditional/country food and health issues. Therefore, the organization of the data gathered here will be used to further develop a conceptual framework on social, cultural and spiritual benefits of country foods for this purpose. Methods of linking this framework with the existing data and approaches to considering health and economic benefits and risks of country foods will be also be conducted in the second year of the project.

Part B: Development of the survey tool

The data collected in this year of the project allowed for the identification of the salient beliefs and preferences regarding traditional/country food and store-bought food within the study population. Knowing the many possible personal influences on food choices of different groups of the population allowed us to build a community-based questionnaire and to test it in one region (Nunavik). Arrangements have been made to apply this survey in the fall of 2000, to give a quantitative assessment of these determinants of food choices. The same process will be repeated in one other region (funded under another program) to verify the survey tool for its cross-cultural applicability.

Deliverables

Deliverables produced in this project will include:

1. Final conceptual framework to understand the social, cultural and spiritual benefits of traditional/country foods and traditional/country food-related activities;
2. Methods for linking this information with the quantitative based health and economic benefits and risks associated with traditional/country foods in the North;
3. Results, analysis and interpretation of application of the survey tool in case communities (i.e. which factors appear most influential in determining diet choice behaviour in the region); and,
4. Overall framework for balancing information related to health, economic, and socio-cultural benefits and risks of traditional/country foods in the North presented in an easily understandable and accessible format for northern health decision-makers and professionals.

Expected Project Completion Date

March 2001

Acknowledgments

We would like to thank Mohan Denetto and Marie-Anick Maille for their work on the literature review and analysis of data on the social, cultural and spiritual benefits of traditional/country foods in this project.

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Risk Assessment for Dioxin-Like PCBs

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Abstract

Two PCB (polychlorinated biphenyl) mixtures were developed, based on residue analysis from Arctic marine mammal fat samples (beluga and narwhal), and tested separately or in combination for their ability to induce a variety of dioxin-specific biochemical and toxicological effects. PCB mixture I contained 9 "dioxin-like" PCB congeners (77, 81, 126, 169, 105, 114, 118, 156, 157) while PCB mixture II contained 15 of the most prevalent nondioxin-like congeners (44, 49, 52, 74, 84, 87, 95, 99, 101, 110, 138, 149, 153, 180, 187). In rat hepatoma cells (H4IIE), TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin) as the positive control, and PCB mixture I induced EROD (7-ethoxyresorufin O-deethylase) activity in a dose dependent manner at a similar potency as described by toxic equivalency factors. PCB mixture II, even at concentrations up to five orders of magnitude higher than TCDD, did not induce EROD activity. PCB mixture I, when combined with TCDD, had limited effect outside of additivity while PCB mixture II had limited apparent effect on the ability of TCDD to induce EROD activity. In vivo induction of hepatic EROD activity was also detected in female B6C3F1 mice given oral doses of both TCDD and PCB mixture I for 14 consecutive days. These results imply that although marine mammals usually contain limited amounts of dioxins and furans, the more abundant PCB fraction does possess dioxin-like activity and this should be taken into consideration when formulating risk:benefit advice for country food consumers. Experiments are continuing to determine possible organ histopathological effects and endocrine disruption potential of the PCB mixtures.

Objectives

1. To determine if PCBs found in country foods possess "dioxin-like" activity.
2. To determine if nondioxin-like PCBs in country foods affect any potential "dioxin-like" activity of other contaminants.

Introduction

Polyhalogenated aromatic hydrocarbons, such as polychlorinated dioxins, furans and biphenyls (PCDDs, PCDFs, PCBs), comprise a group of human-made persistent contaminants which can be found in all ecosystem compartments. Dioxins, furans and PCBs enter the environment as complex mixtures of distinct isomers, each unique based on chlorine substitution patterns. Although these two classes of chemicals share numerous structural, physical and chemical properties, the dioxins and furans are generally considered to be less volatile than the PCBs and therefore less likely to be subjected to temperature-dependent long-range atmospheric transport.

The greater volatility of PCBs is reflected in results from temporal monitoring programs available from certain Great Lakes, once considered to be highly polluted. Water and sediment sample analysis from Lake Michigan indicates yearly decreases in total reserves of PCBs of up to 1600 kg, mainly through atmospheric volatilization (Pearson et al. 1996). These annual declines in PCBs are also reflected in lower PCB body burdens in the top aquatic species such as lake trout. From 1974 to 1990, PCB concentrations in Lake Michigan lake trout declined by almost an order of magnitude (20 $\mu g \cdot g^{-1}$ to 2-3 $\mu g \cdot g^{-1}$) (Feeley and Jordan 1998). Similar drastic PCB declines have also been observed in lake trout collected from Lake Huron over the period of 1976 to 1994 (8.07 $\mu g \cdot g^{-1}$ to 0.47 $\mu g \cdot g^{-1}$) (Scheider et al. 1998).

Conversely, temporal monitoring of ringed seals collected at Holman, Northwest Territories, between 1972 and 1991, has indicated no declines in blubber PCB concentrations since 1981 (Addison and Smith, 1998). Ringed seal and narwhal collected from Lancaster Sound during the 1980s and 1990s have also shown no decreases in PCB concentrations. This is consistent with the concept that historical use patterns combined with physico-chemical properties of certain persistent organic pollutants results in long-range transport to northern climates, with the Arctic acting as a storage reservoir or "sink" (Arctic Monitoring and Assessment Programme 1998).

In an attempt to standardize and facilitate the risk assessment of dioxins and furans, the concept of toxic equivalency factors (TEFs) was developed. Briefly, the potency of each "dioxin-like" compound is compared to the most toxic congener, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), and then assigned a numerical "ranking" or TEF. Each detected congener is then multiplied by its TEF value and all the products summed to give a single TCDD toxic equivalency (TEQ). Inherent in the use of TEFs is that the toxicological mechanism of action for all the congeners is mediated by a common element, the aryl hydrocarbon (Ah) receptor and that interactions between various congeners can be explained by simple additivity. TEFs for dioxins and furans have undergone extensive experimental validation, and their use in risk assessment has been adopted by the majority of human health agencies, including Health Canada, United States Environmental Protection Agency and the World Health Organization (WHO).

Experimental evidence has indicated that certain of the PCB congeners which bioaccumulate in environmental media, including humans, also possess dioxin-like toxicological properties relevant to human health (vitamin depletion, hormonal disruption, reproductive-developmental and immune toxicity, tumour promotion, etc.) (Safe 1994). Unlike dioxins and furans, detailed knowledge of sources and pathways leading to food chain accumulation of dioxin-like PCBs appears to be poorly defined (Alcock et al. 1998). In order to account for the possible dioxin-like activity of PCBs, WHO has suggested that the use of TEFs be extended for initially up to 13 PCB congeners. The human health risk for dioxin would therefore now be comprised of both a component that traditionally originates from the dioxins and furans plus an additional component from a select group of PCBs ($\text{TEQ} = \Sigma[\text{dioxin}_i \times \text{TEF}_i] + \Sigma[\text{furan}_i \times \text{TEF}_i] + \Sigma[\text{PCB}_i \times \text{TEF}_i]$) (van den Berg et al. 1998).

Initial evaluations have indicated that the potential additional dioxin risk due to PCBs differs between the terrestrial and aquatic food chains. Bioconcentration and metabolic capability of marine mammals in particular results in an increased capacity to accumulate certain potential toxic PCBs compared to land mammals (Kannan et al. 1988). For southern populations dependent upon land mammals as major sources of dietary protein and fat, the overall contribution that PCBs would make to a dioxin TEQ would be minimal (Ryan 1997). Conversely, for northern populations dependent upon fish-eating sea mammals, adjusting the TCDD TEQ value for PCBs could result in an increase of up to 300%. This could potentially result in health hazard assessment advice limiting the consumption of traditional country food items such as seal, beluga, walrus and narwhal, known to be good sources of energy (calories), essential fatty acids and fat-soluble vitamins (Kuhnlein 1995).

Recent analysis of contaminants present in the serum of Great Lakes fish eaters indicates that dioxins and furans would account for approximately 60% of a total TCDD TEQ, with the nonortho-substituted PCBs (non-PCBs) accounting for approximately 18% and monoortho-substituted PCBs (mono-PCBs) accounting for approximately 22% (Cole et al. 1997). In comparison, a total TCDD TEQ value from Inuit plasma samples collected from Nunavik residents would comprise a 20% contribution from dioxins and furans, 18% from non-PCBs and approximately 62% from mono-PCBs (Ayotte et al. 1997). Total dioxin TEQs were elevated almost seven-fold compared to either southern Quebec residents or Great Lakes fish eaters, due primarily to the contribution from PCBs. A similar TEQ distribution pattern has also been found in residents of coastal communities along the north shore of the St. Lawrence River basin who consume large quantities of country foods (Ryan et al. 1997).

Higher body burdens of persistent organic pollutants such as dioxins and PCBs in Arctic residents has been attributed mainly to the consumption of sea mammal fat (Dewailly et al. 1993). Overall, it can be concluded that Arctic residents who consume sea mammals could be exposed to potentially greater amounts of TCDD TEQs and have higher body burdens of TCDD TEQs than southern residents, mainly due to the contribution from dioxin-like PCBs.

However, the use of PCB TEFs for risk assessment has not been unconditionally accepted by regulatory and scientific communities, based in part on the possibility of potentiation and/or inhibition of dioxin toxic responses by related food-borne contaminants, including the nondioxin-like PCBs (Safe 1998). We therefore decided to investigate the potential dioxin activity of PCB mixtures commonly found in Arctic marine mammals, and whether or not the more prevalent nondioxin-like PCB fraction could affect dioxin responses.

Activities in 1999-2000

All PCB congeners were obtained as high purity standards (99%+) from a commercial supplier (AccuStandard, Inc.) and analyzed by high resolution gas chromatography-mass spectroscopy for the possible presence of contaminating dioxins, furans or PCBs.

PCB mixtures I and II (Table 1) were constructed based on published reports of PCB congener distributions in Arctic beluga and narwhal. In vitro EROD activity in rat hepatoma cells was conducted as per Sanderson et al. (1996). For the in vivo experiments, female B6C3F1 mice were administered various doses of TCDD, PCB mixture I, PCB mixture II or combinations of the three by gavage on a daily basis for 14 days. Doses were based on a starting total PCB concentration of 4 $\mu g \cdot g^{-1}$ fat and a consumption estimate of up to 100 $g \cdot day^{-1}$ for narwhal blubber. On Day 15, 24 hours after the final dose, each mouse was anaesthetized with isoflurane (Janssen, Toronto, Ontario), exsanguinated and necropsied. Organ weights were recorded for liver, kidney, thymus and brain. Tissues and organs were then processed or stored. Liver microsomes were prepared and cytochrome P450 activity in liver microsomal fractions monitored using a variety of alkoxyphenoxazone substrates (ethoxy, methoxy, benzyloxy and pentoxy [Lubet et al. 1990]). Microsomal protein concentrations were determined (Lowry et al. 1951), using bovine serum albumin as a standard. Enzyme activity was determined by linear regression analysis and reported as nmoles substrate hydrolysed per mg protein per minute. Statistical differences were identified by analysis of variance (ANOVA) followed by Tukey's multiple range testing using SigmaStat (Jandel Scientific, San Rafael, California). When tests for normality or equal variance failed, a non-parametric method (Kruskal-Wallis) was used. Differences were considered significant at $p < 0.05$.

Results and Discussion

Based on the relative distribution of PCB congeners found in marine mammal blubber, and as indicated in Table 1, an estimated dioxin equivalent dose of approximately 70 pg TCDD could conceivably be ingested per gram of blubber. Ingestion of a single serving of blubber (up to 100 g) would therefore result in exceeding the current Health Canada tolerable daily intake (TDI) for dioxins by almost 10-fold. While temporary excursions above the TDI may be considered safe, more frequent consumption patterns would result in higher tissue or organ concentrations, which could ultimately reach a toxicity threshold.

For PCB mixture I, it appears that a "dioxin-like" response, similar to that with TCDD alone, can be generated in hepatoma cells (Figure 1). These in vitro results are in agreement with recent experiments using an environmentally relevant mixture of dioxins, furans and dioxin-like PCBs in the congener mass ratios found in food where relative potency for enzyme induction and immunotoxicity could be explained by TEFs (DeVito et al. 2000). Adding varying concentrations of PCB mixture I in the presence of TCDD increased the amount of detectable EROD activity (Figure 2), while preliminary results also indicated that the standard Ah receptor-mediated EROD induction by TCDD was not affected by a mixture of nondioxin-like PCBs (Figure 3), which on a congener mass basis represents the majority of the total PCB fraction. In support of these findings, a complex mixture of dioxins, furans and PCBs extracted from Great Lake trout has been shown to induce early life stage mortality in fertilized fish eggs similar in potency to what would be predicted by applying TEFs (Wilson and Tillitt 1996).

Table 1. PCB congener make-up of PCB mixture I and PCB mixture II

PCB Congener (IUPAC No.)[a]	% of Congener in Total PCBs in Blubber Sample[b]	Amount of Congener in Blubber Sample[b] ($pg \cdot g^{-1}$ lipid)* ($ng \cdot g^{-1}$ lipid)**	TEF[c]	TEQ[d] (pg TCDD[e])
PCB mixture I				
77	0.0054	216*	0.0001	0.02
81	0.0015	60*	0.0001	0.006
126	0.005	200*	0.1	20.0
169	0.0044	176*	0.01	1.76
105	1.24	49.6**	0.0001	4.96
114	0.625	25**	0.0005	12.6
118	3.81	152.4**	0.0001	15.2
123	ND[f]			
156	0.5	20**	0.0005	10.0
157	0.25	10**	0.0005	5.0
167	ND[f]			
189	ND[f]			
Total PCB TEQ (mixture I)				**69.5**
PCB mixture II				
44	2.0	80**	0	0
49	4.0	160**	0	0
52	10.0	400**	0	0
74	1.8	72**	0	0
84	2.52	94.69**	0	0
87	2.7	108**	0	0
95	7.5	300**	0	0
99	6.0	240**	0	0
101	8.1	324**	0	0
110	4.10	164.0**	0	0
138	9.53	381.2**	0	0
149	6.88	275.2**	0	0
153	10.28	411.2**	0	0
180	2.0	80.0**	0	0
187	3.06	122.4**	0	0
Total PCB TEQ (mixture II)				**0**
Total PCB congeners in blubber sample	**86.7**	**3469.7****		

[a] International Union of Pure and Applied Chemistry

[b] Based on 4 μg total PCBs in a 1 g lipid sample of narwhal blubber

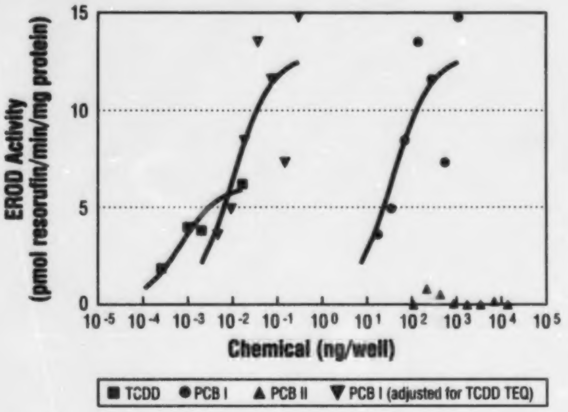
[c] Toxic equivalency factor

[d] TCDD equivalents

[e] 2,3,7,8-tetrachlorodibenzo-*p*-dioxin

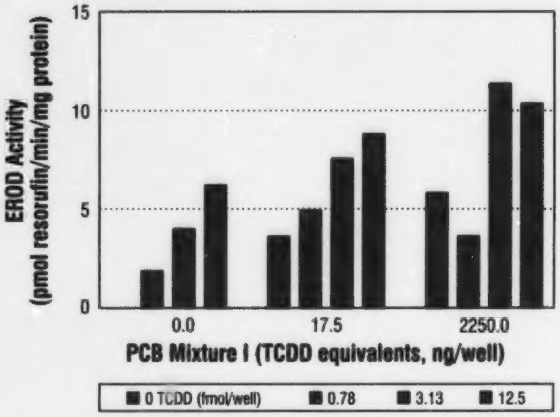
[f] Not detected (Although considered to be dioxin-like, PCB congeners 123, 167 and 189 are not generally detected in arctic marine mammal fat samples and therefore were not included in PCB congener make-up of PCB Mixture I.)

Figure 1. H4IIE EROD activation by TCDD and PCB Mixtures*



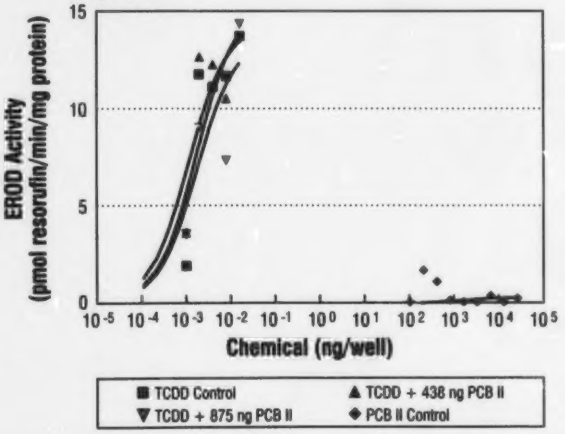
* 18-hour incubation

Figure 2. Effect of PCB Mixture I on H4IIE EROD induction by TCDD*



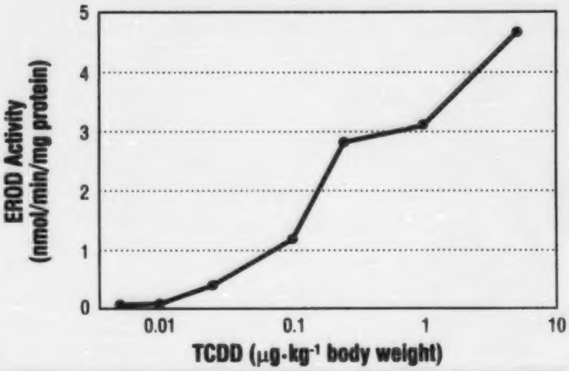
* 18-hour incubation

Figure 3. Effect of PCB Mixture II on H4IIE EROD induction by TCDD*



* 18-hour incubation

Figure 4. Hepatic EROD induction in female mice



While PCB 153, a prominent nondioxin-like PCB congener, has been shown to alter hepatic distribution patterns of other dioxin-like contaminants in rats, no effect was seen on EROD induction (van der Plas et al. 1998). Initial results for the in vivo portion of the study are currently in the data analysis stage except for the control TCDD-EROD induction curve (Figure 4).

Conclusions

Environmental contaminants such as dioxins and furans are known to cause a variety of adverse health effects in humans, including developmental impairment, cancer, diabetes, birth defects and endocrine disruption. Experimental evidence also indicates that exposure to certain PCBs may be associated with similar effects based on a "dioxin-like" contribution. Attempts to define the nature of this potential dioxin response from PCBs have focussed on the use of TEFs (van den Berg et al. 1998).

Based on residue analysis from Arctic Inuit populations, the country food supply is not expected to be highly contaminated with dioxins and furans. However, results from Inuit breast milk screening programs conducted in 1989-1990 did indicate higher concentrations of PCBs (up to five-fold) when compared to southern populations (Gilman 1997). Subsequent dietary surveys have shown that certain populations in the Canadian Arctic can ingest up to 20-fold higher amounts of PCBs on a daily basis as compared to southern populations, with marine mammals being the primary sources of exposure (Kinloch et al. 1992). Furthermore, if the PCB residues found in Inuit breast milk are converted to dioxin "equivalents" using TEFs, the body burden could approach those possibly associated with neurodevelopmental effects in human infants.

The preliminary results of this study provide evidence that PCBs commonly detected in marine mammals are capable of inducing a "dioxin-like" response. Based on the limited dioxin/furan residue analysis available, it would appear as though PCBs could account for the majority of potential dioxin-like activity. The implications for PCB risk assessment would be that both a dioxin-like risk and a separate PCB risk should be taken into consideration when health authorities offer benefit:risk advice to consumers of these country foods.

Ongoing work is attempting to i) define additional dioxin-like biochemical and toxicological endpoints (endocrine, immunology, body and organ weights, analysis of clinical chemistry data, histology and immunohistochemistry of liver); and ii) further characterize the potential for interaction between TCDD/PCB mixture I and PCB mixture II by molecular techniques (gene and protein analysis of CYP1A1, CYP1A2, and CYP2B9/10 in mouse livers via reverse transcriptase-polymerase chain reaction and Western blotting).

Expected Project Completion Date

December 2000

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Nutrient Benefits of Traditional/Country Food Consumed by Inuit

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Abstract

Food items frequently consumed by Inuit, as determined during a dietary evaluation study, were sampled and analyzed for several nutrients. Reported here are selected analyses prioritized by food and nutrient, and the existence of earlier data. Most of these values reported here are first reports of these nutrients in these foods. Nutrient data from this study were incorporated into a database used for nutrient analysis of dietary records. Of particular note in these data are high levels of vitamin C in several animal tissues, and first reports of folic acid in Inuit traditional/country food. Excellent sources of vitamin A (retinol), vitamin E (tocopherol), several essential minerals and the n-3 fatty acids are also noted in selected food items. These data contribute substantially to the existing traditional/country food composition database. Knowledge of nutrient contents in Inuit traditional/country food is important for nutrition education initiatives of the Inuit, and for understanding global food diversity.

Objectives

1. To provide new knowledge of nutrients in traditional/country food frequently consumed by Inuit.
2. To support the food sample collection and analyses from the project, Assessment of Dietary Benefit:Risk in Inuit Communities.
3. To conduct analyses of vitamins in the Health Protection Branch lab when the techniques are not available at Centre for Indigenous Peoples' Nutrition and Environment (CINE).

Introduction

The Human Health Blueprint of the Northern Contaminants Program recognizes that benefit:risk assessment must include nutrient information on food consumed. Arctic diets are composed of traditional and market food items, and while most market foods have known values for the spectrum of nutrients in human nutrition, the complete nutrient profiles of traditional food items are not available. Contrary to popular belief, there are many nutrients in Arctic food for which nutrient levels are not known or are not completely known. This information is needed in order to make informed benefit:risk evaluations of the pattern of food use by residents of Inuit communities, weighing contaminant risks with multiple nutrient and sociocultural benefits. Further, nutrient composition of traditional/country food is in demand by nutritionists working with Arctic peoples in the areas of dietary evaluation, preparation of education materials and in individual counseling.

The project, Assessment of Dietary Benefit:Risk in Inuit Communities, identified the traditional food items most frequently consumed in representative Inuit communities, and team members of that project were active in taking samples of this food as consumed. Methods for analyses of several nutrients were developed during the previous year, and continued during this project. Nutrient analyses of food samples were conducted for proximate composition (protein, fat, moisture, ash, carbohydrate-by-difference, and computed energy), fatty acids (polyunsaturates, monounsaturates, saturates, sum of n-6 and n-3 fatty acids), minerals (calcium [Ca], iron [Fe], copper [Cu], zinc [Zn], phosphorus [P], magnesium [Mg], manganese [Mn], potassium [K], sodium [Na], selenium [Se]), and vitamins A (retinol), E (alpha tocopherol), C (ascorbic acid), folic acid, B-2 (riboflavin) and B-6 (pyridoxine). The method for vitamin D was not yet successfully developed, and results for this vitamin are therefore not included in this report. The Health Canada Health Protection Branch (HPB) laboratories of Drs. Peace and Hidiriglou contributed data of several vitamins. This report contains results of the nutrient analyses for the most frequently consumed Inuit traditional food items that are new information to the CINE nutrient database.

Activities in 1999-2000

The first activity was to prioritize samples for analyses of each nutrient, since it was prohibitive to analyze all samples for all nutrients. Prioritizing was done by reviewing the rank-ordered list of food reported in the 24-hour recalls (n = 1930). Items reported consumed by ≥5 of the randomly selected adults (n = 58 food items) were included in the prioritized list. Second, food items from this list without any nutrient data and for which samples were in hand were given priority. A final prioritization was made for each nutrient in each of the rank-ordered foods according to expectation of whether or not it would be a likely source of the nutrient (e.g. blubber or other pure fat samples were not analyzed for protein, minerals or the B vitamins).

Food samples were portioned for each nutrient and stored appropriately (e.g.as homogenates for vitamins, as freeze dried powders for minerals, as whole tissue for labile vitamins, etc.). Samples for the HPB lab were shipped on dry ice. Throughout this work, sea mammal mattak, a food with both blubber and skin, was dissected and analyzed separately as blubber or as skin (noted as mattak/skin). This was done because of the difficulties in assuming a consistent component of blubber, and the very different nutrient contents of the two portions. This process was carried through in the dietary analysis reported in a companion paper in this volume ("Assessment of Dietary Benefit:Risk in Inuit Communities (Phase III").

Nutrients were analyzed using standard methods (Helrich 90, Behrens and Madere 1994, Hidiroglou et al. 1998, Hoppner and Lampi 1989, Hoppner and Lampi 1993, Kuhnlein and Soueida 1992, Kuhnlein et al. 1991, Tamura 1998, Thompson and Hatina 1979, Esteve et al. 1998, Horne and Patterson 1988, a modification of Fellman et al. 1982). Data were compiled as unit·$100g^{-1}$ food as consumed for ease of insertion in the dietary analysis database.

Results

Results of prioritized analyses as noted above in 10 to 18 frequently consumed food by Inuit are presented in Tables 1 to 5. It was not always possible to ensure a large number of samples of prioritized food items due to delays in dietary data processing. However, the data compiled were adequate for preparing the nutrient database for processing the dietary data with respect to nutrient benefits. Pre-existing data is not included in these summaries but were used to complete the database. Samples included in this report vary in each of the tables due to prioritizing of analyses for each group of nutrients. Results from additional samples are not included in these tables but are available upon request.

In Table 1, new data are presented for proximate composition and energy. Of particular note are the high fat, low protein and resulting high energy levels for fat samples (beluga blubber, narwhal blubber, caribou fat). Meat samples have expected levels of protein, with special note of dried caribou which has higher amounts of protein due to moisture loss. Tables 2a and 2b present values for nutritional minerals per 100 g of edible portion (fresh weight). Of note are the high mineral levels in dried

Table 1. New data for proximate composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Proximate Composition and Energy Values per 100 g*					
			Moisture	Crude Fat	Protein	Ash	Carbohydrates	Energy
Species and Part	Preparation	n	(g)	(g)	(g)	(g)	(g)	(kcal)
Arctic char, flesh	raw	3	77.91 ± 5.38	2.43 ± 3.22	18.50 ± 2.53	1.17 ± 0.19	0.00	96 ± 38
Arctic hare, flesh	boiled	2	62.54 ± 2.00	4.13 ± 1.00	32.47 ± 0.83	0.86 ± 0.17	0.00	167 ± 12
Beluga, mattak/skin	raw	10	69.17 ± 2.52	6.03 ± 2.94	21.48 ± 2.22	0.84 ± 0.28	2.57 ± 2.26	150 ± 24
Beluga, blubber	raw	7	12.76 ± 14.10	70.40 ± 14.31	9.33 ± 8.79	0.22 ± 0.23	7.29 ± 8.16	700 ± 126
Beluga, oil	aged	1	0.23	98.33	0.33	0.00	1.11	891
Blackberries**	raw		—	—	—	—	—	—
Caribou, fat	raw	1	2.63	89.91	1.15	0.01	6.30	839
Caribou, flesh	boiled	2	63.72 ± 1.82	4.77 ± 1.63	30.61 ± 0.22	0.77 ± 0.15	0.13 ± 0.18	166 ± 15
Caribou, flesh	dried	2	20.29 ± 11.96	5.37 ± 2.60	68.46 ± 9.88	3.07 ± 0.49	2.81 ± 1.02	333 ± 59
Caribou, flesh	raw	2	71.45 ± 0.57	3.10 ± 1.13	23.05 ± 0.07	1.20 ± 0.07	1.20 ± 1.70	125 ± 3
Cranberries**	raw		—	—	—	—	—	—
Muskox, flesh	raw	2	75.25 ± 1.94	2.37 ± 2.87	19.40 ± 2.30	1.40 ± 0.29	1.58 ± 2.23	105 ± 6
Narwhal, blubber	raw	2	9.87 ± 2.68	79.59 ± 15.70	8.03 ± 11.18	0.14 ± 0.18	2.37 ± 2.01	758 ± 88
Narwhal, mattak/skin	raw	5	71.18 ± 2.26	5.27 ± 2.04	20.91 ± 1.93	0.96 ± 0.06	1.67 ± 0.77	138 ± 19
Polar bear, flesh	boiled	1	54.32	2.94	41.16	1.58	0.00	191
Ptarmigan, flesh	boiled	1	62.38	4.20	32.39	1.02	0.00	167
Ringed seal, flesh	boiled	2	78.10 ± 17.28	1.64 ± 1.31	19.03 ± 16.84	0.90 ± 0.40	0.33 ± 0.47	92 ± 77
Ringed seal, flesh	raw	4	69.03 ± 1.86	2.22 ± 1.96	19.51 ± 12.82	1.28 ± 0.21	7.97 ± 14.25	130 ± 16
Walrus, blubber**	aged, raw	1	32.13	58.11	2.77	0.30	6.69	561
Walrus, flesh	aged, raw	1	64.05	10.85	24.03	1.08	0.00	194

* Mean ± standard deviation

** Not analyzed (existing data)

caribou meat, high Fe in meats in general, especially seal, and high Zn and Na in mattak/skin.

Table 3 presents new data for vitamins A (retinol) and E (tocopherol). Beluga blubber, aged beluga oil, caribou liver, narwhal blubber and ringed seal liver are shown to be excellent sources of both nutrients. The variability in results from multiple samples of the same species and part are likely due to dietary sources of the animal. Nevertheless, these are recognized as excellent sources of vitamins despite the variability noted.

Table 4 presents values in selected samples for the water-soluble vitamins riboflavin (B-2), total pyridoxine (analyzed as three forms), folate and ascorbic acid. Several excellent sources of these nutrients are identified. For example, a 100 g portion of caribou liver or ringed seal liver would meet or exceed the adult daily recommended intake for riboflavin, pyridoxine and folate. Excellent sources of ascorbate are beluga mattak/skin, caribou liver, narwhal mattak/skin, and ringed seal brain and liver.

In Table 5, new data for fatty acids are presented for 10 selected food items. Data are presented as the sum of polyunsaturated fatty acids (PUFA) which includes the sum of the n-3 and n-6 fatty acids (n-3 sums are for 18:3, 20:3, 20:5, 22:3 and 22:6) (n-6 sums are for 18:2, 18:3, 20:2, 20:3, 20:4, 22:2 and 22:4). Monounsaturated fatty acids (MFA) include the sums of 12:1, 14:1, 16:1, 18:1 oleate, 18:1 vaccinate, 20:1 5-eicos, 20:1 11-eicos, 22:1 and 24:1. Saturated fatty acids (SFA) include the sums of 8:0, 10:0, 12:0, 14:0, 16:0, 18:0, 20:0, 22:0 and 24:0. These fatty acid summations are compatible with earlier results of fatty acid contents of Arctic traditional food. Excellent sources of PUFA and n-3 fatty acids are cisco and burbot roe, eider duck eggs, aged beluga oil and burbot liver.

Table 2a. New data for mineral composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Mineral Concentration per 100 g*				
			Ca	Fe	Cu	Zn	Mg
Species and Part	Preparation	n	(mg)	(mg)	(mg)	(mg)	(mg)
Arctic char, flesh	raw	4	13.3 ± 8.7	0.4 ± 0.1	0.05 ± 0.02	0.3 ± 0.1	35.0 ± 11.7
Arctic hare, flesh	boiled	1	9.5	6.7	0.55	2.6	38.1
Beluga, mattak/skin	raw	10	5.1 ± 1.4	0.4 ± 0.1	0.05 ± 0.01	6.8 ± 0.7	15.6 ± 1.8
Beluga, blubber	raw	6	1.1 ± 0.7	0.1 ± 0.1	0.02 ± 0.02	0.1 ± 0.1	0.9 ± 0.9
Beluga, oil**	aged		—	—	—	—	—
Blackberries	raw	7	4.9 ± 0.9	0.2 ± 0.2	0.10 ± 0.04	0.1 ± 0.03	5.3 ± 0.3
Caribou, fat**	raw		—	—	—	—	—
Caribou, flesh	boiled	1	3.9	5.5	0.28	7.6	26.3
Caribou, flesh	dried	2	22.1 ± 21.0	10.5 ± 0.9	0.73 ± 0.04	12.0 ± 2.1	78.9 ± 2.8
Caribou, flesh	raw	6	3.6 ± 1.3	5.5 ± 2.4	0.35 ± 0.10	2.9 ± 0.9	33.5 ± 11.8
Cranberries	raw	4	11.0 ± 5.5	0.2 ± 0.1	0.09 ± 0.03	0.2 ± 0.1	6.5 ± 0.9
Muskox, flesh	raw	1	3.2	4.5	0.13	2.4	25.1
Narwhal, blubber	raw	2	1.2 ± 0.3	0.3 ± 0.1	0.03 ± 0.03	0.3 ± 0.4	0.8 ± 0.1
Narwhal, mattak/skin	raw	3	5.6 ± 1.9	0.3 ± .04	0.04 ± .001	8.2 ± 1.5	16.3 ± 1.7
Polar bear, flesh	boiled	1	7.1	7.0	0.30	12.7	35.1
Ptarmigan, flesh	boiled	1	45.3	7.3	0.48	1.8	37.6
Ringed seal, flesh	boiled	1	2.5	27.9	0.10	3.1	31.8
Ringed seal, flesh	raw	5	3.3 ± 1.0	24.1 ± 2.5	0.22 ± 0.16	2.5 ± 0.5	30.5 ± 2.0
Walrus, blubber**	aged, raw		—	—	—	—	—
Walrus, flesh	aged, raw	1	5.9	22.0	0.15	5.4	27.1

* Mean ± standard deviation

** Not analyzed (low priority)

Discussion and Conclusions

The results of this project give new and exciting nutrient results, particularly for vitamins, in Inuit traditional/country food items. These data are compatible with earlier reports on nutrient levels, and contribute substantially to the nutrient composition database. These data also make a valuable contribution to benefit:risk assessment on the use of Inuit traditional/country food, as it becomes more clear that these food items should be protected and included in the diet whenever possible. Computer modeling of benefit and risk can proceed with a better understanding of a more complete nutrient profile in contrast to contaminant content profiles of traditional food items. Additionally, there is still much to be learned about the disease prevention potential of these nutrients in the Inuit diet and how they may contribute to the mediation of contaminant health effects. Continuing research is needed to address additional nutrients, particularly vitamin D and niacin (B-3), and to expand the sample size to understand nutrient variability in similar tissues of species.

As noted in other reports, the vast array of traditional food resources of Indigenous peoples must be documented because of their important place in global nutrition and food diversity (Burlingame 2000, Kuhnlein 2000. These important sources of nutrients are receiving new attention from many international, national and regional agencies and governments. Traditional food resources of the Inuit have a significant place in this knowledge because of the excellent sources of nutrients contained in a food system primarily comprised of animal food species.

For the Inuit health education network, it is necessary to recognize these sources of nutrients in order to conduct

Table 2b. New data for mineral composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Mineral Concentration per 100 g*				
			Na	K	P	Mn	Se
Species and Part	Preparation	n	(mg)	(mg)	(mg)	(mg)	(mg)
Arctic char, flesh	raw	4	68.5 ± 50.7	515.7 ± 86.3	231.5 ± 53.3	0.004 ± 0.003	bdl***
Arctic hare, flesh	boiled	1	34.3	263.2	208.0	0.031	bdl
Beluga, mattak/skin	raw	10	95.6 ± 20.7	326.3 ± 33.5	132.5 ± 17.3	0.003 ± 0.002	0.24 ± 0.19
Beluga, blubber	raw	6	16.0 ± 5.6	3.8 ± 6.0	6.0 ± 3.1	bdl	bdl
Beluga, oil**	aged		—	—	—	—	—
Blackberries	raw	7	2.9 ± 1.2	99.3 ± 14.0	7.3 ± 0.4	1.411 ± 0.707	bdl
Caribou, fat**	raw		—	—	—	—	—
Caribou, flesh	boiled	1	37.3	261.6	162.2	0.030	bdl
Caribou, flesh	dried	2	84.1 ± 21.7	1166.7 ± 17.5	539.2 ± 21.0	0.069 ± 0.003	0.02 ± 0.02
Caribou, flesh	raw	6	36.3 ± 13.4	451.8 ± 118.1	206.1 ± 59.1	0.027 ± 0.007	bdl
Cranberries	raw	4	3.0 ± 1.4	89.8 ± 13.7	7.6 ± 1.7	4.666 ± 4.312	bdl
Muskox, flesh	raw	1	50.9	419.8	159.2	0.012	bdl
Narwhal, blubber	raw	2	24.5 ± 1.6	15.0 ± 2.3	7.6 ± 2.8	0.005 ± 0.003	bdl
Narwhal, mattak/skin	raw	3	102.0 ± 31.2	369.0 ± 29.7	155.0 ± 15.5	0.002 ± 0.001	0.44 ± 0.26
Polar bear, flesh	boiled	1	59.5	551.9	245.8	0.019	0.07
Ptarmigan, flesh	boiled	1	37.9	249.3	221.8	0.033	0.02
Ringed seal, flesh	boiled	1	34.6	303.3	186.5	0.007	0.06
Ringed seal, flesh	raw	5	37.6 ± 4.5	426.3 ± 50.3	206.5 ± 30.7	0.016 ± 0.009	0.03 ± 0.01
Walrus, blubber**	aged, raw		—	—	—	—	—
Walrus, flesh	aged, raw	1	66.1	320.9	142.5	0.043	bdl

* Mean ± standard deviation

** Not analyzed (low priority)

*** Below detection limit

meaningful education and counseling activities for Inuit to enhance the health promotion agenda.

Expected Project Completion Date

March 31, 2001

Acknowledgments

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Table 3. New data for fat soluble vitamin composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Vitamin Concentration per 100 g*	
Species and Part	**Preparation**	**n****	**Retinol (mg)**	**Tocopherol (mg)**
Arctic char, flesh	raw	3	0.03 ± 0.03	0.19 ± 0.05
Arctic hare, flesh	boiled	3	0.04 ± 0.05	0.39 ± 0.19
Bearded seal, intestine	boiled	2	0.04 ± 0.03	1.72 ± 0.29
Beluga, mattak/skin	raw	7	0.27 ± 0.09	1.16 ± 0.39 (10)
Beluga, blubber	raw	10	2.37 ± 1.95	4.84 ± 3.76
Beluga, oil	aged	1	2.03	29.05
Burbot, eggs	raw	4	0.09 ± 0.03	3.52 ± 1.94
Caribou, fat	raw	1	0.06	0.68
Caribou, flesh	boiled	1	0.01	0.54
Caribou, flesh	raw	4	0.12 ± 0.15	3.52 ± 0.29
Caribou, liver	raw	5	6.27 ± 2.68	12.64 ± 11.32
Narwhal, blubber	raw	6	1.90 ± 1.79	8.92 ± 11.22
Narwhal, mattak/skin	raw	6	0.31 ± 0.31	1.42 ± 0.35
Polar bear, flesh	raw	1	0.43	0.27
Ptarmigan, flesh	boiled	1	0.05	0.49
Ringed seal, flesh	boiled	2	0.01 ± 0.01	0.27 ± 0.07
Ringed seal, flesh	raw	5	0.12 ± 0.19	0.09 ± 0.05
Ringed seal, liver	raw	3	3.71 ± 3.04	3.64 ± 3.61
Walrus, blubber	aged, raw	1	0.19	0.59
Walrus, flesh	aged, raw	1	0.04	0.16

* Mean ± standard deviation

** Unless indicated otherwise in parentheses

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Table 4. New data for water soluble vitamin composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Vitamin Concentration per 100 g*			
Species and Part	**Preparation**	**n****	**Riboflavin (mg)**	**Pyridoxine (mg)**	**Folate (mg)**	**Ascorbate (mg)**
Arctic char, flesh	raw	1	0.02	—***	—	1.23 ± 0.03 (2)
Arctic hare, flesh	boiled	1	0.46	0.87	0.01	—
Bearded seal, intestine	boiled	2	0.20 ± 0.10	1.17 ± 0.13	0.05 ± 0.02	6.12 ± 4.90
Beluga, mattak/skin	raw	3	0.02 ± 0.005 (2)	0.80 ± 0.005	bdl†	36.02 ± 8.73 (6)
Blackberries	raw	1	0.01	—	0.01 ± 0.004 (4)	2.41 ± 0.80 (5)
Caribou, kidney	boiled	2	2.10 ± 0.78	1.51 ± 0.27	0.08 ± 0.07	7.24 ± 3.14 (4)
Caribou, liver	raw	5	1.58 ± 0.48	1.97 ± 0.62	0.31 ± 0.20	23.76 ± 4.92 (4)
Caribou, stomach	raw	1	0.48	1.01	—	0.45 ± 0.39 (6)
Caribou, tongue	raw	1	0.13	0.50	bdl	—
Cranberries	raw	2	0.02 (1)	0.25 ± 0.01	0.02 ± 0.01	3.88 ± 1.30
Narwhal, mattak/skin	raw	2	0.03 ± 0.01	0.75 ± 0.21	—	31.51 ± 6.96 (6)
Polar bear, flesh	raw	1	0.28	0.79	0.01	—
Ptarmigan, flesh	raw	2	0.51 ± 0.10	1.63 ± 0.23	0.03 ± 0.04	1.70 (1)
Ringed seal, brain	raw	1	0.11	0.50	0.02	14.86 ± 2.60 (4)
Ringed seal, flesh	boiled	1	0.22	0.93	0.02	1.19
Ringed seal, flesh	raw	2	0.27 ± 0.03	1.16 ± 0.37	—	1.55 ± 0.50 (3)
Ringed seal, liver	raw	3	1.10 ± 0.19	2.20 ± 0.28	1.12 ± 0.12	23.80 ± 3.82
Walrus, flesh	raw	2	0.13 ± 0.06	0.52 ± 0.14	0.02 ± 0.003	1.00 (1)
Walrus, kauk	raw	1	0.05	0.27	—	0.70
Walrus, flesh	aged, raw	1	0.08	0.44	0.01	0.85

* Mean ± standard deviation

** Unless indicated otherwise in parentheses

*** Not analyzed (low priority)

† Below detection limit

Table 5. New data for fatty acid composition of selected frequently consumed Inuit traditional foods, listed alphabetically

Food Sample			Fatty Acid Concentration per 100 g*				
			PUFA**	n-3	n-6	MUFA***	SFA†
Species and Part	Preparation	n	(g)	(g)	(g)	(g)	(g)
Arctic cisco, eggs	raw	2	3.30 ± 2.94	3.16 ± 2.85	0.14 ± 0.10	6.10 ± 1.73	3.98 ± 2.53
Arctic hare, flesh	boiled	1	1.22	0.08	1.14	1.02	2.00
Beluga, oil	aged	1	14.89	13.41	1.48	51.97	18.59
Burbot, eggs	raw	2	3.81 ± 0.92	3.16 ± 1.05	0.65 ± 0.12	3.33 ± 1.30	0.83 ± 0.13
Burbot, liver	raw	2	2.55 ± 2.62	1.91 ± 2.23	0.63 ± 0.40	6.48 ± 2.25	4.00 ± 0.02
Caribou, flesh	aged	1	0.53	0.02	0.51	0.93	1.22
Caribou, kidney (pooled)	boiled	2	1.78 ± 0.24	0.15 ± 0.03	1.63 ± 0.20	1.39 ± 0.97	2.25 ± 1.14
Caribou, liver	raw	3	0.46 ± 0.19	0.05 ± 0.01	0.42 ± 0.19	0.44 ± 0.17	1.26 ± 0.57
Eider duck, eggs	boiled	1	1.17	0.46	0.71	7.74	4.88
Ringed seal, brain	raw	1	0.82	0.60	0.22	0.91	1.19

* Mean ± standard deviation

** Polyunsaturated fatty acids

*** Monounsaturated fatty acids

† Saturated fatty acids

Assessment of Dietary Benefit:Risk in Inuit Communities (Phase III)

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Abstract

This was the third and final year of the project to document estimates of traditional and market food use among adult Inuit in five regions of the Canadian Arctic. This year's activities included data entry, laboratory analyses of traditional food samples, data analyses, and discussion of the results in a workshop with regional Inuit representatives. Results indicate contaminants of greatest mean intake and concern to be chlordane, toxaphene and to a somewhat lesser extent, mercury. Intakes varied by age, gender and region. Nutrient benefits are substantial and are often derived from the same food species and parts in which elevated levels of contaminants are found. On days when Inuit consumed traditional food, their nutrient intakes for several nutrients were greater than on days without traditional food. Cultural, economic and social benefits of traditional food resources are also documented to be great for all regions and communities. These dietary evaluation results parallel those found earlier in one Baffin community, however, the variability across regions for traditional food use, contaminant exposure and nutrient intake from traditional food is considerable. A companion article in this volume (Nutrient Benefits of Traditional/Country Food Consumed by Inuit) and forthcoming reports for this project provide additional and more extensive information. Communication initiatives that will bring results of this project to Inuit communities in 2000-2001 are being coordinated by the Inuit Tapirisat of Canada.

Objectives

This was the third and final year of the project, which had the overall objectives:

1. To derive quantitative estimates of traditional and market food intake among adult Inuit in five regions (Inuvialuit, Kitikmeot, Keewatin, Baffin and Labrador), representing approximately 50 Inuit communities.
2. To complete databases of nutrient and contaminant contents of traditional food as prepared and consumed for quantitative estimates of intake of these items.
3. To define benefits of traditional food in terms of nutritional, socioeconomic and cultural significance.

4. To define the levels of dietary exposure to contaminants (mercury [Hg], cadmium [Cd], arsenic [As], lead [Pb] and several organochlorines).

During this final year of the project, the objectives were:

1. To complete interviews and food sample collection.
2. To complete dietary data entry.
3. To complete laboratory analyses for nutrients and contaminants.
4. To complete dietary data analysis.
5. To present a draft report to a workshop of representatives from the five Inuit regions for discussion, interpretation and guidance.
6. To prepare the project report.

Introduction

The Northern Contaminants Program (NCP) Human Health Blueprint outlines priorities that include dietary evaluation for contaminant exposure, and evaluation of both benefits and risks of traditional/country food use. This project was developed based on the Centre for Indigenous Peoples' Nutrition and Environment (CINE) experience and methodology developed with similar NCP projects in 26 communities in Denendeh and the Yukon. Similar methods were used to derive annual estimates of benefits and risks in one Inuit community in Baffin Island (Kinloch et al. 1992, Kuhnlein 1995, Kuhnlein et al. 1995a, Kuhnlein et al. 1995b, Kuhnlein et al. 1996, Chan et al. 1995, Chan et al. 1997). The need to understand food use benefits and risks in Inuit communities in several regions was identified by the NCP in 1996. It was anticipated that the types, as well as quantities, of traditional/country food used and the benefits and risks derived would vary across the major Inuit regions. Dietary evaluations give the advantage of understanding which food items contribute various proportions of the nutrient or contaminant under review, and how alternate sources of nutrients can be obtained while reducing intake of contaminants within the framework of existing dietary patterns (Kuhnlein and Receveur 1996).

Estimates of dietary exposure to simultaneous multiple contaminants have been identified, and these exposures are of concern. Meanwhile, concerns also relate to the consequences of a shift toward increased consumption of market food, a shift that may be accelerated by knowledge of the presence of contaminants in traditional/country food. Since traditional/country food is recognized as being rich in nutrients and cultural importance, it is important to balance information about contaminants in traditional/country food with information on the various benefits of its use, including the nutrient properties of the food as well as the social and cultural benefits that the use of traditional/country food provides.

During the first year of this project (1997-1998), the research methods and processes were defined, and potential collaborators were identified. Databases of existing information on nutrients and contaminants in traditional/country food species were compiled and information gaps were identified (Kuhnlein et al. 1991, Kuhnlein and Soueida 1992, Chan 1998). Five regional workshops were held from October 1997 to February 1998, to which 39 Inuit communities sent representatives. Each workshop identified the extent of traditional/country food species used seasonally by communities in that region (total species n = 194, all regions), and recommended the communities in which to conduct the dietary evaluation research. The overwhelming interest in this study meant that while all community representatives wished the evaluation to take place in their community, a selection had to be carefully made by workshop participants, based on factors such as the diversity of the local food system, community size, existing health concerns and budget limitations of the project. Therefore, the research was to be conducted in 18 communities: Aklavik, Tuktoyaktuk, Paulatuk, Kugluktuk, Holman, Cambridge Bay, Resolute Bay, Pond Inlet, Igloolik, Kimmirut, Baker Lake, Chesterfield Inlet, Rankin Inlet, Nain, Hopedale, Makkovik and Rigolet. It was not possible to conduct the project in one identified community, Pelly Bay, due to budget constraints. An additional budget portion was obtained for the research to be conducted in Qikiqtarjuaq.

During the second year of the project (1998-1999), research agreements were negotiated with each community identified at the workshops, and pilot testing of the interview tools was completed in Shingle Point and Chesterfield Inlet. Research staff were hired and trained at workshops at CINE in Ste-Anne-de-Bellevue, Quebec, in September 1998 and January 1999. Random 10% samples of resident households in the research communities were selected during each time period, and interviewing proceeded with a maximum of two adults and one adolescent from each household. Collection of food samples thought to be of primary importance during the interview period were made. The interviewing periods extended from October to December 1998, and January to early April 1999. Survey instruments included 24-hour recalls, reports of traditional food frequency of use in the prior 3 months, 7-day traditional food use records during the season of interview, a short socioeconomic questionnaire, and height and weight measures of interviewees/participants. During the winter interview period, additional information was gained with repeat

24-hour recalls (n = 172) and a qualitative exercise on food attributes (126). Community food store prices and availability of selected items for a standard market basket of food was completed by research assistants. All data were returned to CINE for computer file entry and processing. In all, 1930 interviews were completed and approximately 200 samples of food prepared for consumption were collected.

Activities in 1999-2000

During this final phase of the project, laboratory and data analyses proceeded at CINE, as did discussion of results in a draft report prepared for a workshop (March 2000) of representatives from participating Inuit communities in each of the five regions.

Laboratory analyses were conducted and data were added to the existing database for four heavy metals (As, Cd, Hg and Pb) and eight major groups of organochlorines (CHL [chlordane], (CBz [chlorobenzene], DIE [aldrin and dieldrin], DDT, HCH [hexachlorocyclohexane], MIR [mirex], PCBs [polychlorinated biphenyls] and TOX [toxaphene] (Chan 1998, Kuhnlein et al. 1991, Kuhnlein and Soueida 1992). Nutrient analyses conducted in the CINE laboratory included proximate composition (energy, protein, fat, carbohydrate-by-difference, moisture, ash), fatty acids, essential minerals (iron [Fe], zinc [Zn], copper [Cu], manganese [Mn], magnesium [Mg], calcium [Ca], phosphorus [P], sodium [Na], potassium [K], selenium [Se]), and vitamins (A [retinol], E [tocopherol]). Health Canada laboratories conducted analyses of selected additional vitamins (ascorbic acid, pyridoxine, folic acid, riboflavin). The rationale for prioritizing food samples for nutrient analyses, as well as results for the most frequently used Inuit foods, are presented in a companion paper in this volume ("Nutrient Benefits of Traditional/ Country Food Consumed by Inuit"). Following the laboratory work, the nutrient and contaminant databases were compiled without missing values for processing of the dietary data.

The 24-hour recall dietary data were treated with the above two databases, additional interview information was summarized, and tables were prepared as a comprehensive draft report. The report was presented to a workshop held March 23-24, 2000 in cooperation with the Inuit Tapirisat of Canada (ITC). Workshop participants included 16 Inuit leaders and representatives of Inuit organizations, representatives of ITC and CINE researchers.

Results

For the purpose of this report, only the highlights of the study will be presented. Further depth and detail is available in the comprehensive report, *Assessment of Dietary Benefit: Risk in Inuit Communities*, prepared as a discussion document for the communication initiatives of the ITC during 2000-2001, and available by request from CINE and ITC.

It is clear that traditional/country food as currently consumed (n = 139 different food items recorded) contains multiple benefits, both for nutrient intake and social-economic-cultural value. This same food also contains multiple contaminants. The contaminants of greatest concern when comparing intake to the current established tolerable daily intake levels (TDI) or provisional tolerable daily intake levels (PTDI) are the organochlorines, chlordane and toxaphene. Table 1 shows the population distribution of intake of heavy metals and organochlorines for all regions combined. Mean intakes of CBz, DDT, HCH, mirex and PCBs were below the TDI/PTDI used by Health Canada, whereas mean intake for chlordane and toxaphene were four times higher than the TDI. The high intake

Table 1. Population distribution of heavy metal intake and organochlorines ($\mu g \cdot kg^{-1}$ body weight$\cdot day^{-1}$) (n = 1875)

	TDI/PTDI* ($\mu g \cdot kg^{-1} \cdot d^{-1}$)	n > PTDI	n > 0	Mean	Median	95th Percentile	95th/PTDI
As	2	209	477	0.6	0.0	4.2	2.1
Cd	1	126	1085	0.3	0.1	1.2	1.2
Hg	0.71	381	1077	0.6	0.1	3.0	4.2
Pb	3.57	433	1063	2.1	0.2	8.8	2.5
CHL	0.05	305	1067	0.2	0.0	0.5	9.4
CBz	0.27	72	1071	0.1	0.0	0.2	0.7
DDT	20	2	1088	0.3	0.0	1.1	0.1
HCH	0.3	42	696	0.0	0.0	0.1	0.5
MIR	0.07	44	377	0.0	0.0	0.0	0.3
PCBs	1	118	1090	0.4	0.0	1.7	1.7
TOX	0.2	381	1074	0.9	0.0	2.2	10.9

* Tolerable daily intake/Provisional tolerable daily intake

sub-group (95 percentile) had 10 times higher exposure of chlordane and toxaphene than the TDI. Mean intake levels of the four heavy metals were below the TDI/PTDI.

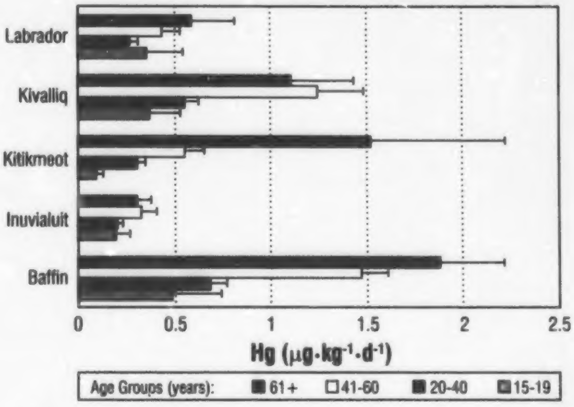
The 24-hour recalls represent one day's intake with two seasons being represented. Most records demonstrated intake of contaminants, and there were many one-day's intakes that exceeded the TDI/PTDI. Those one day's intakes that exceeded the PTDI by more than four-fold, on average, are considered contaminants of concern. Thus Hg (4.2-fold), in addition to chlordane (9.4-fold) and toxaphene (10.9-fold), are in this category.

Overall intake of three groups of organochlorines in the Baffin Region by food species (including all parts of animals) is shown in Table 2. It can be seen that the primary contributors of chlordane, PCBs and toxaphene are narwhal and walrus blubber (60% to 70% of total), with a lesser contribution from beluga blubber. Other species contribute small amounts, although as a whole they represent approximately 30% of intake. These data consider both the content of the food samples as well as the amount of that food consumed.

There are differences in intake by age as well as by region, as shown in Figures 1 and 2. It can be clearly seen that mean intake of older individuals (> 40 years old) contributing 24-hour recalls was greater for Hg than younger individuals in all five regions. Intakes of chlordane, toxaphene and Hg are shown to be greatest in the Baffin region.

Table 3 shows that intake of selected nutrients varies when traditional/country food is or is not consumed. Days with traditional/country food have significantly more energy, protein, n-3 fatty acids, retinol (vitamin A), tocopherol (vitamin E), Fe and Zn. With respect to other benefits of traditional/country food, Table 4 shows the percentage agreement on selected attributes by region. Usually, > 90% of those interviewed agreed that these attributes, which were identified and confirmed in the year 1 workshops, were provided by traditional/country food. In Table 5, results of rank score summaries of traditional/country food and market food demonstrated that interviewees recognized traditional/country food as being significantly more healthy for children, healthy for pregnant/breastfeeding women, tasty and important to

Figure 1. Mean daily intake of Hg in different regions assessment of dietary benefit in Inuit communities*



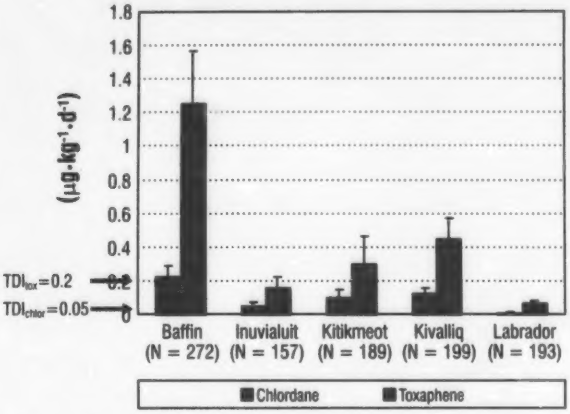
* Mean ± SE

Table 2. Sources of organochlorines in the diet of Inuit in the Baffin Region (contribution %)

Species	Part	Weight	CHL	PCBs	TOX
Caribou	flesh	38.2	0.9	1.3	0.1
Ringed seal	flesh	18.7	0.8	2.4	8.9
Arctic char	flesh	15.6	2.2	1.5	3
Narwhal	muktuk/skin	5	1.8	7	0.1
Walrus	flesh	3.2	1.7	0.6	0.1
Ringed seal	broth	2.9	0.2	1.1	0.1
Polar bear	flesh	2.8	1.5	3.1	0.1
Narwhal	blubber	1.9	37.9	44.5	35.6
Ptarmigan	flesh	1.3	0	0	0
Beluga	muktuk/skin	1.2	1.7	1.7	09
Walrus	blubber	1.2	34.9	22.2	43.1
Beluga	blubber	0.4	11.1	8.5	6.3
Ringed seal	blubber	0.3	1.9	1.3	0.3
Polar bear	fat	0.1	2.3	1.6	0.5
TOTAL (%)		**92.8**	**98.9**	**96.8**	**99.1**

community life. Thus, it is clear from this research initiative that traditional/country food provides multiple nutrient and cultural benefits to Inuit.

Figure 2. Mean daily intake of organochlorines in different regions (Age = 20 to 40 years)*



* Mean ± SE

Table 3. Mean nutrient intake on days with and without traditional food for Inuit women aged 20 to 40 years

Nutrient	Intake on Days with TF* Mean ± SE	Intake on Days without TF Mean ± SE
Energy** (kcal)	2167 ± 60	1824 ± 56
Protein** (g)	155 ± 4	69 ± 4
SFA (g)	24 ± 1	25 ± 1
n-3 (omega) fatty acids** (g)	3 ± 0.1	1 ± 0.1
Vitamin A** (µg)	438 (378, 507)†	301 (262, 346)†
Vitamin E** (mg)	6 ± 0.3	4 ± 0.2
Calcium (mg)	422 ± 19	391 ± 18
Iron** (mg)	36 ± 2	11 ± 2
Zinc** (mg)	22 ± 0.7	9 ± 0.7
Total Sugars	100 ± 5	109 ± 5

* Traditional food

** Significantly different between days with and without TF, $p < 0.001$

† Geometric means plus confidence interval

Table 4. Percentage agreement on selected attributes of traditional foods, by region

	Regions				
Harvesting and using traditional food by the family	**Inuvialuit (n = 335)**	**Kitikmeot (n = 296)**	**Kivalliq (n = 303)**	**Baffin (n = 434)**	**Labrador (n = 353)**
	%	%	%	%	%
Contributes to physical fitness and good health	96	96	98	96	95
Is a favourite outdoor recreation activity	93	94	89	87	93
Provides people with healthy food	98	98	98	98	99
Keeps people "in tune with" nature	96	95	87	91	96
Favours sharing in the community	96	96	95	94	93
Saves money	97	95	94	87	91
Is an essential part of the culture here	99	99	97	96	98
Is an occasion for adults to display responsibility for their children	99	98	93	90	96
Is one way to practice spirituality	76	78	57	57	71
Contributes to humility	86	85	71	62	74
Brings respect from others	97	91	83	89	88
Builds one's pride and confidence	97	90	84	91	97
Provides education on natural environment	96	95	93	91	95
Contributes to children's education	95	96	94	93	96
Provides skills in survival	99	98	97	97	98
Provides skills in food preparation at home	99	98	98	95	98
Is an opportunity to teach spirituality	77	75	61	58	70
Is an opportunity to learn patience and other personality qualities	96	93	83	91	96
Is a way to strengthen the culture	97	95	89	93	98

Table 5. Mean ranking[§] of traditional and market food items by region according to four food preferences criteria

Region	Food type	Healthy for children	Healthy for pregnant/ breastfeeding women	Taste/ like to eat	Important to community life
Inuvialuit	TF[†]	2.4	2.2	2.9*	2.0
(n = 30)	MF[‡]	2.2	2.2	2.6	2.3
Kitikmeot	TF	1.9**	1.7**	2.2	1.5***
(n = 15)	MF	2.5	2.4	2.4	2.1
Kivalliq	TF	2.0*	1.8**	2.3	1.7**
(n = 25)	MF	2.3	2.2	2.3	2.1
Baffin	TF	1.6***	1.4***	1.9***	1.6***
(n = 39)	MF	2.2	2.3	2.6	2.4
Labrador	TF	2.0**	2.0	2.3	1.6
(n = 17)	MF	2.6	2.5	2.5	2.2
All regions	TF	2.0**	1.8***	2.3*	1.7***
(n=126)	MF	2.3	2.3	2.5	2.3

[§] Rank score coding: 1 = most; 5 = least

[†] Traditional food

[‡] Market food

t-test for difference in ranks: *$p < 0.05$, **$p < 0.01$, ***$p < 0.001$

Discussion and Conclusions

This report presents the highlights of an extensive 3-year study of traditional/country food and market food used by Inuit in five regions of the Canadian Arctic. For more in-depth reports, the reader is referred to forthcoming reports and peer-reviewed publications. Communication initiatives will bring results of this project to residents of Inuit communities in 2000-2001 and are being coordinated by the ITC, in cooperation with CINE.

The primary new finding of this study is that the various Inuit regions consume differing proportions of contaminants depending upon the traditional/country food consumed, and that of these five regions, respondents in the Baffin Region consume more of the assessed contaminants. This finding was anticipated due to the consumption of significant amounts of sea mammals food items by Inuit in the Baffin Region. These results are compatible with earlier reports from one community, Qikiqtarjuaq, in the Baffin region. The organochlorines of greatest intake and concern are chlordane and toxaphene, and the heavy metal of greatest intake and concern is Hg. These particular contaminants were identified in earlier reports (Gilman et al. 1997).

Intakes of chlordane and toxaphene at these levels have unknown effects on human physiology. Effects of these levels of Hg intake are similarly unknown, as are the effects of intakes of multiple simultaneous contaminants from the many food species in the traditional food of Inuit. Clearly, more research on the consequences of these levels of intake are needed (Van Oostdam et al. 1999, Kuhnlein and Chan 2000).

As shown in this and the companion report in this volume from CINE, most traditional food items identified as being frequently consumed and containing contaminants, are also good or excellent sources of essential nutrients. For example, sea mammal blubber and mattak/skin are excellent sources of retinol, tocopherol and ascorbic acid, nutrients which are limited in the mix of traditional/country food and market food consumed in Inuit communities. Unless nutrient supplements are routinely used, a decrease in use of sea mammal foods will place some individuals at health risk from nutrient deficiency.

Variations in traditional/country food and market food intake by age and gender show that young women, usually considered most at risk from contaminants due to the potential for pregnancy, are consuming less traditional/ country food than are older adults, and are at the lower end of the distribution of population intake. It follows that they are also likely to be at the lower end of the distribution of nutrient intake. Further data management and research will clarify these issues.

Traditional food use by Inuit in the regions assessed provides multiple cultural, social and economic benefits, in addition to substantial nutrient benefits. The effects of losing these benefits as a result of decreasing use of traditional/country food are of concern in several ways, including increased incidences of diabetes and heart disease, and effects on mental health (Gilman et al. 1997). This message must be used to greatest advantage to spur the efforts of environmental protection of Arctic ecosystems and control of global contaminant emissions.

Project Completion Date

March 31, 2000

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Transplacental Exposure to PCBs and Infant Development/Human Exposure Assessment

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Abstract

The objective of this study is to examine the consequences of in utero and lactational exposure to polychlorinated biphenyls on Inuit infants, from birth to 12 months of age. Of particular interest is the impact of exposure on newborn thyroid hormone levels and physical growth, on the infant's overall health, mental, psychomotor and neurobehavioural development, and on visual and spatial information processing abilities. The data collection of this ongoing study started in November 1995 and will end in 2001. The continuation of data collection was the main activity carried out in 1999-2000. The data collection in Greenland began in March 2000. Communication activities were performed in Nunavik communities. To date, 229 Inuit mothers in Nunavik have completed the prenatal interview and 146 have completed the postnatal interview; 127 infants were assessed at 6.5 months and 122 were assessed at 11 months of age. There are no preliminary results available regarding the effects of exposure to polychlorinated biphenyls, and effects analysis will wait until the final sample size of 300 11-month-old infants (200 from Nunavik, 100 from Greenland) is obtained.

Objective

To examine the consequences of in utero and lactational exposure to PCBs (polychlorinated biphenyls) on Inuit infants from birth to 12 months of age.

Introduction

Results from several surveys have indicated that Inuit newborns from Nunavik (northern Quebec) are exposed to doses of organochlorines which exceed acceptable (tolerable) daily intakes for several health endpoints. The mean concentration of PCBs (Aroclor 1260) in breast milk fat was 2.9 $mg \cdot kg^{-1}$ ($n = 107$) for Inuit women, compared with 0.5 $mg \cdot kg^{-1}$ ($n = 536$) for women from southern Quebec (Dewailly et al. 1992). This level among Inuit is 5.6 times greater than that of southern women and is > 1.5 $mg \cdot kg^{-1}$, the lipid concentration corresponding to the maximum acceptable concentration in breast milk of 50 $\mu g \cdot L^{-1}$ established by Health Canada guidelines (Health Canada 1978). More recently, PCB concentrations were measured in Greenlandic newborns. The average PCB (Aroclor 1260, fat basis) concentration measured in cord blood samples was 1.7 $mg \cdot kg^{-1}$ (P. Bjerregaard, unpublished data), compared with 1.0 $mg \cdot kg^{-1}$ in Nunavik (Dewailly et al. 1995) and 0.2 $mg \cdot kg^{-1}$ in southern Quebec (Rhainds et al. 1999).

The effects of prenatal exposure to background levels of PCBs from environmental sources have been examined in four major cohort studies: in Michigan, North Carolina, the Netherlands and New York. The effects of PCB/PCDF (polychlorinated dibenzofuran) poisoning were also studied in Taiwanese children. In Michigan, higher cord serum PCB concentrations were associated with lower birth weight (Fein et al. 1984). This exposure was also associated with less optimal newborn behavioural function (e.g. reflexes, tonicity and activity levels) in all three studies that administered the Brazelton Neonatal Behavioral Assessment Scale (Jacobson et al. 1984, Lonkey et al. 1995, Rogan et al. 1986). The Netherlands study found that elevated levels of PCBs can alter the thyroid hormone status in early infancy; higher thyroid-stimulating hormone (TSH), and lower free thyroxine (FT_4) and total thyroxine (TT_4) levels were found to be significantly correlated to PCBs measured in maternal milk (Koopman-Esseboom et al. 1994). In North Carolina, dose-response analysis indicated deficits in psychomotor development at 6, 12 and 24 months in highly exposed children, i.e. those whose mothers' milk PCB levels exceeded 1.75 $mg \cdot kg^{-1}$ (Gladen et al. 1988, Rogan and Gladen 1991). At higher levels of PCB/PCDF exposure in Taiwan, deficits were seen in infant mental and psychomotor development (Yu et al. 1991). The major finding in Michigan was an association with poorer visual information processing ability, which indicated a deficit in visual memory and/or attention (Jacobson et al. 1985, Jacobson et al. 1990, Jacobson et al. 1992).

The research protocol of the longitudinal study and laboratory methods have been described previously (Muckle et al. 1999a).

Activities in 1999-2000

Data collection of this ongoing longitudinal study started in November 1995 and will end in August 2001. The continuation of data collection was the main activity carried out in 1999-2000. We experienced difficulties obtaining the cord blood samples of participants during 1999-2000 because of lack of collaboration with the midwives responsible for blood sampling at delivery. However, in cases when the cord blood sample was missing, we were successful in obtaining the maternal blood sample. Since the correlation between cord blood PCBs and maternal blood PCBs concentrations is very high (Aroclor 1260 on fat basis, $r = 0.972$, $p < .001$, $n = 81$), this will not have a significant impact on the methodological quality of the research. Numbers of samples collected and analyzed in 1999-2000 are presented in Table 1.

To date, 229 mothers have completed the prenatal interview and 146 have completed the postnatal interview; 127 infants were assessed at 6.5 months and 122 were assessed at 11 months of age. The recruitment slowed down slightly in 1999-2000 because we had not anticipated that 12% of the new declared pregnancies would involve mothers who had already enrolled in the study with a previous child and who were not eligible to participate in the study again. Numbers of maternal interviews performed and infants tested during 1999-2000 are presented in Table 2.

Table 1. Number of samples collected and analyzed in 1999-2000

Maternal hair samples	49
Cord blood samples	17
Maternal blood samples	44
Infant blood samples at 6 month of age	43
Breast-milk samples	34

Table 2. Number of maternal interviews performed and infants tested during 1999-2000

Prenatal interviews	33
Postnatal interviews	22
Infants tested at 6.5 months of age	25
Infants tested at 11 months of age	37

The data collection in Greenland began in March 2000.

A poster and pamphlet were released in the communities where the research takes place (Puvirnituq, Inukjuaq, Kuujjuaraapik). These materials informed the population about the objectives and procedures used in this study. In addition, Gina Muckle did a phone-in radio program at the local FM stations in Puvirnituq and Inukjuaq, which provided an update to the population on the research activities carried out since September 1997 and information about what will be done during the following year in their community.

Gina Muckle and Carole Vézina met with the newly elected mayors of Puvirnituq and Inukjuaq in order to fully explain to them the objectives and the procedures of this study. Meetings were also held with new hospital and nursing station nurses, midwives and laboratory technicians directly involved in the data collection process to present the objectives of the research and the research procedures. Further meetings were held with all medical professionals involved in the study so that problems and difficulties emerging since the beginning of the data collection could be identified.

A new research assistant was enrolled and was trained in Québec and in Detroit to perform maternal interviews and infant testing in the field.

Results

Preliminary exposure data and participation rates were presented previously (Muckle et al. 1999a, Muckle et al. 1999b). There are no preliminary results available regarding the effects of exposure to PCBs on infant health and development. Effects analysis will wait until the final sample size of 300 is obtained. The updated participation and refusal rates are presented in Table 3.

Since a prospective study involving a large number of Inuit women in Nunavik of reproductive age and their children has the potential to address issues of concern for community and health authorities, we met with municipal councils, community groups and health authorities and made a commitment to bring back to them any information we can in the course of the study. With respect to this, we are working on two brief reports to be presented to public health authorities and the Nunavik Nutrition and Health Committee. The first one focuses on the sociodemographic and dietary determinants of exposure to PCBs and mercury (Hg) and the second one addresses the issue of the determinants of psychological distress in Inuit women in Nunavik. The abstracts of these two reports are presented below.

Table 3. Participation and attrition rates, up to March 31, 2000

Declared pregnancies (n = 333)	**333**	
Exclusion: Already in study (45/333)	45	13.5%
Unable to contact (7/288)	7	2.4%
Initial refusal rate (52/281)	52	18.5%
Initial participation rate (229/281)	229	81.5%
*Eligible for postnatal interview (n = 179)**		
Exclusion:		
Moved to another village (6/179)	6	3.4%
No biological sample (10/179)**	10	5.6%
Baby adopted in an other village (5/179)	5	2.8%
Miscarriage and perinatal mortality (8/179)	8	4.5%
Unable to contact (1/150)	1	0.7%
Refusal rate (3/149)	3	2.0%
General attrition (33/179)	33	18.4%
Participation rate (146/149)	146	98.0%
Eligible for 6.5 month assessment (n = 140)†		
Exclusion:		
Medical condition	1	0.9%
Moved to another village	8	5.7%
Died	1	0.7%
Unable to contact (4/131)	4	3.1%
Refusal rate (0/127)	0	0.0%
General attrition (13/140)	13	9.3%
Participation rate (127/127)	127	100%
Eligible for 11-month assessment (n = 122)‡		
Exclusion: Moved to another village	0	0.0%
Unable to contact	0	0.0%
Refusal rate (2/122)	2	1.6%
General attrition (2/122)	2	1.6%
Participation rate (120/122)	**120**	**98.4%**

* 50/179 women had not delivered yet and therefore were not eligible for the 1-month postnatal interview.

** No biological sample had been taken at birth; therefore, the infant was excluded.

† 6/146 infants had not reached the age of 6.5 months yet and therefore were not eligible for testing.

‡ 7/127 infants were tested at 6.5 months of age but had not yet reached the age of 11 months and therefore were not eligible for testing.

Sociodemographic and dietary determinants of exposure to PCBs and mercury in Inuit women in Nunavik

This study identified maternal characteristics associated with country food consumption and documented traditional food items associated with an increased PCBs and Hg body burden in pregnant Inuit women from northern Quebec. Pregnant women were invited to participate in a study of infant health and development. Country food consumption and demographic characteristics were documented from interviews conducted at mid-pregnancy and 1 and 12 months postpartum. PCB concentrations were measured in plasma; Hg concentrations were measured in whole blood and hair; and selenium (Se) concentrations were measured in whole blood. Women reported eating significant quantities of fish, beluga fat, seal meat and seal fat. Maternal demographic characteristic (including level of education, number of adults in household, crowding in the home, number of cigarettes smoked daily, and likelihood of giving their baby up for adoption) were good predictors of fish consumption during pregnancy. By contrast, maternal demographic characteristics were not predictors of beluga and seal consumption. Surprisingly, fish and marine mammal consumption during pregnancy were not associated with blood Hg concentrations, however, they were associated with higher hair Hg concentrations. Fish, beluga and seal consumption during pregnancy were not associated with PCB body burden, and beluga fat was the main source of Se intake during pregnancy. Demographic characteristics might help to identify Inuit women of childbearing age who consume fish frequently.

Determinants of psychological distress in Inuit women in Nunavik

Psychological distress, suicide, alcohol and drug use, and family violence are problems of concern in Nunavik (northern Quebec, Canada). However, the incidence and determinants of these problems are not well documented. The aim of this study is to document the incidence of psychological distress in Nunavik women of childbearing age, and to identify factors associated with psychological distress. Pregnant women ($n = 110$) from three Nunavik communities were invited to participate in an infant development study. Sociodemographic characteristics, French or English literacy, non-verbal reasoning skills, smoking, alcohol, domestic violence, psychological distress (thought problems, anxiety, irritability, depression), and suicidal ideation and attempts were documented from structured interviews conducted at mid-pregnancy and 1 and 12 months postpartum. Socioeconomic status ($r = -.22$) and adaptation to the broader North American culture ($r = .24$) were associated with maternal anxiety and irritability ($p \leq .05$). Domestic violence was most strongly associated with anxiety ($r = .56$) and distress ($r = .50$). Alcohol use before pregnancy was related to depression ($r = .23$, $p \leq .05$) and suicidal attempts ($r = .38$, $p \leq .05$), while alcohol use during pregnancy was related to all the distress indicators and more strongly with depression ($r = .23$ to $.37$, $p \leq .05$). The data point to domestic violence and alcohol use as two critically important factors associated with increased psychological distress in Nunavik women of childbearing age.

Conclusions

There are no results to present at this stage in relation to the study objective. The research and communication activities planned for year 1999-2000 were performed. The participation rate in the present study has been high and very few women have withdrawn their collaboration during the course of the study. In light of the work that has been done to date, no changes have to be made to the activities and time lines for year 2000-2001. At the end of the year 2000-2001, prenatal exposure data will be available for about 230 participants. These data will be used to monitor long-term trends analysis of human exposure. Results will be available for about 165 infants tested at 11 months of age.

Expected Project Completion Date

December 2001

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Follow-Up of Preschool Age Children Exposed to PCBs and Mercury through Fish and Marine Mammal Consumption

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Abstract

This study is designed to examine the long-term neurophysiological and neurological consequences of prenatal exposure to polychlorinated biphenyls and methylmercury. Two pilot projects involving 16 of children were performed in Montreal and Kuujjuaq (Northern Quebec) to pre-test the instruments used for assessment of the visual system and neuromotor, cognitive and attentional functions in preschool age children. Some adjustments had to be made to the selected instruments in order to adapt them to the children's age and attention span limits. The two pilot studies confirmed that the selected instruments, and the tests originally designed for adults, have been successfully adapted for children of preschool age. The pilot studies were quite useful for the testing of ambulatory equipment, determining the feasibility of fine neuromotor testing, and finally, recording visual evoked potentials and cognitive evoked potentials in sub-optimal testing conditions. With the suggested changes, we are confident that the next field trip (to Puvirnituk and Inukjuak) will provide higher quality data and results. There are no results to present at this stage in relation to the study objective.

Objective

To examine the long-term neurophysiological and neurological consequences of prenatal exposure to polychlorinated biphenyls (PCBs) and methylmercury (MeHg).

Introduction

The effects of prenatal exposure to PCBs and MeHg resulting from fish consumption have been studied in fish eating populations from various countries. Prenatal exposure to PCBs and MeHg have been linked to deficits in neurobehavioural development during infancy. However, only two studies provided data for school age children, and neuromotor as well as neurophysiological effects were not fully documented. Although neurophysiological testing was performed in one of these studies with school age children exposed to MeHg, the study remained incomplete. Furthermore, no study has been aimed at investigating cognitive deficits under MeHg exposure using cognitive evoked potentials (ERPs).

The purpose of this research is to examine the long-term consequences of prenatal exposure to PCBs and MeHg. This project is designed to extend previous findings by studying domains of effects that have been overlooked in most of the previous studies (neurophysiological and neurological endpoints) that could be related to learning difficulties and disabilities. The large number of Aboriginal children in Northern Quebec who are highly exposed due to PCBS and Hg due to maternal consumption of fish and marine mammals provides a unique opportunity to: i) study the impact of PCBs and MeHg on child development; ii) provide the information required for the development of blood guidelines, no effect thresholds, acceptable daily intakes, and health advisories for Canadian children and women of reproductive age; and iii) broaden our knowledge on the nature of specific deficits associated with exposure to these environmental contaminants. The sample will comprise 100 Nunavik Inuit children between 5 and 6 years of age. Two subsets of 50 children will be formed according to in utero PCBs and MeHg exposure levels (low and high). Prenatal exposure to PCBs and MeHg was documented from cord blood analysis at birth (Dewailly et al. 1997). Groups will be comparable with respect to socio-economic status. A neurological exam will be performed, and the fine and gross neuromotor function as well as the visual system and the cognitive and attentional functions will be assessed. Potential demographic, socio-familial and biological confounders will be assessed via maternal interviews and blood sample analysis.

Activities in 1999-2000

The Nunavik Cord Blood Surveillance Program database permitted the identification of those children who would participate in the study, as well as their prenatal PCBs and MeHg exposure.

Computerized tests designed to assess fine neuromotor functions in children were developed; tests designed to assess gross neuromotor functions in children were selected; and, apparatus for electrophysiological testing was purchased, assembled and tested. The instruments to document confounding factors were adapted for the Inuit population and administered to approximately 200 Inuit women in another cohort of Inuit women living in Northern Quebec.

This project received approval from several agencies. The Laval University Ethical Committee approved the research protocol and procedures; the Ungava Tulattavik Health Center (Ungava Bay region in Northern Quebec) and the Inuulitsivik Health Centre (Hudson Bay region in Northern Quebec) authorized the study; and municipal councils of Kuujjuaq, Puvirnituk and Inukjuak have approved the study. These three communities will be visited by the research team during the year 2000.

A pilot study was planned in the initial grant proposal and the anticipated completion date of this activity was December 3, 1999. The objective of this first phase was to pre-test the instruments adapted or developed to assess the fine neuromotor function as well as the visual system and the cognitive and attentional functions in preschool age children. This pilot project was performed in Montreal between November 29, 1999 and December 6, 1999.

A second pilot study was planned in the initial grant proposal for February 2000. The objective of this first phase was to pre-test the instruments adapted after the first pilot phase held in Montreal on Inuit children of the targeted age group. This pilot project was performed in Kuujjuaq in February 2000.

Results

Visual evoked potentials

The visual evoked potentials (VEPs) used here are a reflection of the neuromechanism responsible for the spatial integration of the visual information, and allow an appropriate evaluation of the maturation and functional integrity of the visual pathway.

The pilot study done in Kuujjuaq used a VEP protocol which had been revised and compressed following the Montreal pilot study. This revised protocol included two

Table 1. Visual acuity of eight Nunavik Inuit children using Snellen E chart for visual acuity and Titmus test for stereopsis

Subjects	Visual Acuity Right eye	Visual Acuity Left eye	Stereopsis (seconds of arc) Animal test	Circle test
1	20/50	20/50	100	0
2	20/30	20/30	40	40
3	20/30	20/30	100	50
4	20/50	20/50	100	400
5	20/30	20/30	Not done*	Not done
6	20/30	20/30	100	0
7	20/30	20/30	100	Not done
8	20/30	20/30	3 000	0

* Incomplete tests may be due to the lack of interest of the child at the end of the testing period.

spatial frequencies (4 and 8 cycles·degree^{-1}) and variable contrasts 95%, 30%, 20%, 4%, 2% and 1% for 4 cycles·degree^{-1} and 95%, 16%, 4% and 2% for 8 cycles·degree^{-1}. We tested eight children using this VEP protocol along with the visual screening tests using the Snellen E chart for visual acuity and the Titmus test for stereopsis. The visual acuity and the stereopsis tests were well understood and performed by children. Most of the Inuit children had a visual acuity of at least 20/30, which could be considered normal given the testing conditions, which were not up to standard testing conditions in clinical settings. However, two of the eight patients had a subnormal visual acuity of 20/50 in both eyes (subjects 1 and 4) and two others had lower stereopsis scores than expected for their visual acuity (subjects 6 and 8) (Table 1). Moreover, since myopia could be more common in Inuit because of racial factors, as in Asiatic eyes, refractive errors using the stenopeic hole will be screened when visual acuity appears subnormal. When the stereopsis scores are revealed to be low for normal visual acuity, the photograph of the child taken for the neurologic evaluation will be will be used to identify children with strabismus.

All eight subjects were evaluated for all parameters of the revised VEP protocol. For the spatial frequency 4 cycles per degree, five of the eight children were successfully tested with all seven contrasts. The lowest contrast (1%) was too weak to evoke a response in most of the subjects. Among the remaining subjects, the results of two were invalidated by technical problems (unstable electrodes) or artefacts (eye movement), and another one did not cooperate sufficiently to generate valuable data. The absence of reliable responses in the evaluation of these two latter subjects correlated with their poor collaboration as shown by their results in the graded attention scale. Furthermore, the VEP protocol was sometimes performed following the ERP protocol which meant the possibility of an overly long electrophysiological testing time for some children. Moreover, since the type of stimuli used in both ERPs and VEPs were quite comparable (black and white grating versus checkerboard), this could have contributed to the lack of interest from the child. Consequently, fatigue and over-extended attention spans from the combined protocols appeared to be an important limiting factor to obtaining good, reliable data on all children.

For the spatial frequency 8 cycles·degree^{-1}, VEP results were similar to the spatial frequency 4 cycles·degree^{-1}, except that reproducible waves were difficult to obtain for both children with the 20/50 visual acuity (subjects 1 and 4, Table 1), especially for the low contrasts. This could be explained by the low visual acuity and inability to detect such small grating stimuli. The mean latency and amplitude of VEP waves at each contrast were analyzed, but since all normative data are not available, no comparison could be made for the time being (Table 2).

Considering the above, some methodological adjustments were made to the revised protocol to improve the quality of the results in future field trips. First, the testing time was reduced, as it was found to be too long for children tested with the general testing protocol (neuromotor, neurological and cognitive protocols) to maintain their alertness and cooperation. Therefore, we are planning to keep the two spatial frequencies but use fewer contrasts than what was used in the two pilot studies. A reduction in the ERP protocol is also planned. Secondly, the high spatial frequency (8 cycles·degree^{-1}) was decreased to 7 cycles·degree^{-1} to ensure the detection of the stimulus in cases of subnormal visual acuity. Consequently, the low spatial frequency (4 cycles·degree^{-1}) was increased to 3 cycles·degree^{-1} in order to maintain a sufficient difference in the parameter to test separately the two visual pathways (magnocellular and parvocellular). These changes in the parameters will be adequate to obtain the information

Table 2. Spatial frequency 4 cycles per degree (n = 8)

	95%	30%	20%	12%	4%	2%	1%
Latency (msec)							
Mean	108.8	121.9	118.8	121.4	125	128	ND**
SE*	4.87	3.8	4.24	3.83	4.2	1.5	
Amplitude (μV)							
Mean	20.2	13.23	14.8	13.8	18.43	7.85	ND
SE	7.27	10.8	6.0	5.9	4.9	4.2	

* Standard error of the mean

** Non-detectable wave

required to test our hypotheses, while reducing the testing time.

Cognitive evoked potentials

The function of ERPs is to measure changes in the brain electrical activity related to a psychological event or the presentation of a given stimulus. With ERPs we can estimate the levels of vigilance, attention, and semantic processing or even neurological or auditory disorders.

With respect to ERPs, two different procedures had initially been planned: P300 peak (using an oddball paradigm) and Mismatch Negativity (MMN). These procedures will still be used, however during the pilot study in Kuujjuaq, a series of methodological problems were encountered. First, of the eight children tested during the 1-week period, four showed some hearing impairments and two others had visual impairments. Although the two remaining children did not seem physically impaired, their data was highly influenced by artefacts (e.g. eye movements). These artefacts are believed to be linked to low attention levels during testing. A means of increasing attention is to counterbalance the presentation of the different protocols (VER and ERP). Unfortunately, even counterbalancing the VEP and the ERP protocols within the general testing sequence appeared unable to eliminate major sources of artefact. Therefore, this raised concerns over the choice and the administration of stimuli for the auditory and the visual oddball protocols.

First, the hearing impairments (mostly the inability to distinguish tones of different frequencies) were important enough to limit the discrimination of the frequent from the deviant auditory stimuli during the general introductory screening procedures. Second, considering the length of the general testing protocol (neuromotor, neurological and neurophysiological testing add up to more than 3 hours of testing) and children's limited attention span, it appeared important to shorten the cognitive ERP sequence. Shortening the ERP protocols enhances attention in children, diminishes the impacts of artefacts and thus increases the amount of data fit for later analysis.

As such, based on results obtained during the pilot study in Kuujjuaq, two major changes will be made to the ERP protocols. First, the auditory P300 protocol will be removed. The second recording protocol, employing a visual P300 (oddball paradigm) associated with a motor task (reaction time), added to the general ERP protocol following the Montreal pilot study, will still be employed. However, the initial visual paradigm used black and white checkerboards or grey scalings. The choice of those stimuli, although standard when the ERP and VEP protocols are administered independently, seems inappropriate with Inuit children. The attention demands appear to be too elevated for these children when the protocols are jointly administered. Children appeared to find the task to be extremely boring and thus, attention levels were significantly affected. It thus appears necessary to modify the visual stimuli in order to maintain the child's attention and to decrease the possibility of artefact rejection. At the same time, however, task demands should not vary. Frequent stimuli will now consist of 8 cm^2 red squares, while deviant stimuli will consist of blue circles. The frequent stimuli (red square) will be presented with an 80% probability of occurrence while the deviant stimuli (blue circle) will occur on the remaining 20% of trials. The inter-stimulus interval will be fixed and set at 1200 ms. Again, children have to detect the deviant visual stimulus and signal this detection by pressing a button switch. The MMN will be used to quantify the degree of hearing loss in children. In any case, the MMN protocol will still be used. Both protocols have been extensively used in previous research and will be essential to evaluate and test our hypotheses.

Neuromotor functions

Gross motor function

In the original protocol, we had not planned to assess gross motor function. However, we then decided to add this developmental domain to the original protocol since Cordier and Garel (1998) recently found diminished leg coordination in children exposed to MeHg (results not published). In order to discriminate normal and at-risk preschool children on the basis of neurological tests, Huttenlocher et al. (1990) suggested a small, easily performed battery of gross motor performance. From the 24 items of the original battery (Touwen and Prechtl 1970), they recommended 11 tasks that discriminated between normal and at-risk children. This examination is considered to represent sensorimotor screening tests for early detection of preschool children with suspected neurodevelopmental disorders. These tasks include walking on toes, walking on heels, tandem gait (forward and backward), touch localization, restless movement, hand coordination, downward drift, hopping, alternating and complex tapping. The observation of data obtained after the two pilot studies revealed the existence of ceiling and floor effects for some of the previous items. All the children succeeded in the touch localization item and all the children failed in the complex alternating tapping tasks. Thus, these two items were not sensitive enough and have been excluded from the evaluation.

Fine neuromotor assessment

Tables 3 to 5 present the preliminary results for some of the fine neuromotor functions (tremor, postural sway, reaction time) and for cardiovascular functions obtained in the two pilot studies. No statistical analyses have been performed because of the small sample size (only eight subjects in each group). Results for the alternating movement tests and for the rapid pointing movement tests are not yet available.

Maintenance of standing posture, tremor and reaction time have long been associated with assessment of nervous system functions. The CATSYS analysis system (Danish Product Development, Ltd. 1996a), is a computerized and portable system used to record these neuromotor functions. Postural tremor is recorded by a light stylus that contains a biaxial micro-accelerometer (TREMOR analysis system: Danish Product Development, Ltd. 1996b). Five key figures were calculated: median frequency, frequency dispersion, tremor intensity, harmonic index and tremor index (Table 3). Reaction time is assessed using a hand-held switch activated by the thumb with a sound stimulus.

To measure postural sway, a platform containing three orthogonal string-gauge devices is used (SWAY analysis system: Danish Product Development, Ltd. 1996c) Six sway parameters were calculated: mean sway, sagittal sway, transversal sway, sway index, sway area and sway velocity. In the initial evaluation, children's postural sway was assessed in one position on the platform: feet

Table 3. Preliminary results for tremor measured by the accelerometer

	Right hand		Left hand	
Characteristics	**Pilot study (Montreal)**	**Pilot study (Kuujjuaq)**	**Pilot study (Montreal)**	**Pilot study (Kuujjuaq)**
n	8	8	8	8
Median frequency (Hz)	5.918	6.060	6.250	5.597
Frequency dispersion (Hz)	2.850	3.080	3.021	2.707
Tremor intensity ($m \cdot s^{-2}$)	0.241	0.256	0.341	0.262
Harmonic index	1.799	1.792	1.842	1.874
Tremor index	2.885	3.082	4.467	3.211

Table 4. Preliminary results for postural sway measured by the force platform

	Eyes open		Eyes closed	
Characteristics	**Pilot study (Montreal)**	**Pilot study (Kuujjuaq)**	**Pilot study (Montreal)**	**Pilot study (Kuujjuaq)**
n	8	8	8	8
Mean sway (mm)	9.243	8.480	9.471	9.487
Sagittal sway (mm)	4.664	4.827	5.692	5.833
Transversal sway (mm)	6.971	6.040	6.167	6.260
Sway index	89.43	111.07	88.92	83.20
Sway area (mm^2)	441.50	303.47	495.33	558.00
Sway velocity ($mm \cdot s^{-1}$)	18.79	16.00	20.33	25.67

Table 5. Preliminary results for cardiovascular functions measured by PROPAQ 106

Characteristics	Pilot study* (Kuujjuaq)
Blood pressure (mean)	105
Systolic/diastolic (mmHg)	66
Blood pressure (SD**)	4.842
Systolic/diastolic (mmHg)	2.923
Heart rate (mean) (beats·minute^{-1})	97
Heart rate (SD)	13.937

* The cardiac monitoring was not evaluated in the pilot study in Montreal.

** Standard deviation

together (eyes opened and eyes closed). A general observation of the performances in static and dynamic equilibrium tasks revealed that Inuit children seem to use a lot of balance reactions (with arms, trunk and legs) (Table 4). In order to thoroughly study this observation, we decided to introduce more difficult equilibrium tasks with the force platform: balance on one foot, heel to toe balance or tandem balance.

All the tests used for the evaluation of the fine neuromotor functions were originally designed for adult motor assessment. Thus, the tests have all been adapted for the evaluation of children by adding visual or auditory reinforcements or by adapting the procedures (total recording time, position adopted, etc.). These important adaptations were established in order to obtain and keep the interest of children and thus optimize their participation throughout the evaluation. Normative data in adults are available (Després et al. 2000) but the normalization of these tests in children of preschool age does not yet exist. In that respect, we are planning to set up a standardization study with healthy children of ages between 4 and 6 years old.

Cardiac monitoring

Sorensen et al. (1999) recently found a significant effect of blood pressure and heart rate variability in children from the Faroe Islands exposed to prenatal MeHg. In the first protocol, we had not planned to assess cardiovascular function. However, because MeHg exposure can be a potential environmental risk factor for alteration of cardiovascular functions in childhood, we decided to add this new element to the initial evaluation. The monitoring of these functions is carried out by a portable system, the PROPAQ 106 (Protocol Systems Inc.). This apparatus, frequently used in hospitals, consists of the installation of an armband (blood pressure cuff) and three electrodes (electrocardiogram lead sets) on precise places on the torso (upper left, upper right and lower left). It allows the assessment of the heart rate, the blood pressure and the electrocardiograph that are printed throughout the recording (Table 5).

Neurological examination

Preliminary results for the neurological evaluation with Amiel-Tison and Gosselin examination revealed no major neurological dysfunctions in all the tested children. At this stage of the study, no tendency can be observed. During these two pilot studies, this examination has been performed by a pediatrician from Ste-Justine Hospital, Montreal, Quebec.

Discussion and Conclusions

There are no results to present at this stage in relation to the study objective. The two pilot studies performed confirmed that the selected instruments, which are designed to assess the neuromotor and neurophysiological functions, as well as the tests originally designed for adults, have been successfully adapted for children of preschool age. Children were very curious and interested in the different tests. The pilot studies were quite useful for the testing of ambulatory equipment and determining the feasibility of recording VER and ERPs in less than ideal conditions (e.g. unavailability of Faraday room, transport of equipment). The administration of testing procedures was appropriate and relatively feasible with the Inuit children. With the suggested changes adapted for our population, we are confident that the next field trip will provide higher quality data and results. In light of the work that has been done to date, no changes have to be made to the activities and time lines for year 2000-2001.

Expected Project Completion Date

March 2002

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Standard Edible Weights of Harvested Species in the Inuvialuit Settlement Region

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Abstract

A literature and file search was conducted for whole body, carcass and edible weights of species harvested in the Inuvialuit Settlement Region. Standard whole body weights were selected from available data sets according to criteria relating to harvest location, season, type and sample size. Empirical measures or conversion rates were applied as appropriate to establish edible weights, taking into account known factors and practices in the Inuvialuit Settlement Region. Standard edible weight estimates were derived for 11 mammal species, 14 bird species and nine fish species, which are estimated to account for approximately 99% of the total harvest by weight. These weights were applied to 10 years of harvest survey data (1988-1997) to determine mean annual country food production in the Inuvialuit Settlement Region and mean per capita country food availability.

Objectives

1. To provide a screening tool for identifying situations of possible human health concern, using available harvest survey and contaminants data to identify the upper bound of potential contaminant exposure via country food consumption, at a community or regional level.
2. To provide guidance and focus for future diet surveys, if required.

Introduction

Edible weight refers, in a subsistence harvesting context, to the weight of those parts of an animal that are normally eaten as country food. For mammals, the closest equivalent is the weight of the dressed carcass less bone and gristle. Various estimates of edible weights for some species may be found in biological, economic, and archaeological literature, however, they have seldom been derived according to a standard and clearly specified procedure, and many are of uncertain origin or applicability. A previous survey of the literature in the 1970s was done in connection with the James Bay and Northern Quebec land claim agreement (James Bay and Northern Quebec Native Harvesting Research Committee 1976), but this included few Arctic species.

This survey provides estimates of both whole body and edible weights for the 34 primary harvested species in the Inuvialuit Settlement Region (ISR). The estimates take into account variation in whole body weights by year, season, life-cycle stage and geographic location, as well as age and sex composition of the harvest, the relation of whole body weight to carcass or dressed weight, the relation of carcass weight to edible weight and local food preferences. The various concepts are defined (Usher 2000) and applied to the available data.

Activities in 1999-2000

In this new, 1-year project, the project leader developed the study methodology and data collection framework, reviewed a wide range of literature, contacted nearly 20 individuals (mostly biologists) who provided guidance, information and in some cases their own unpublished data sets, developed whole body and edible weight estimates, and applied these to a recently available data set (Fabijan and Usher, unpublished data) of estimated Inuvialuit harvest levels in the ISR.

The selection of mean values for this report gives priority to those data sets:

- from the ISR from populations or stocks normally harvested by Inuvialuit for subsistence (where no such data sets exist, preference is given to adjacency);
- from the same season in which subsistence harvesting normally occurs;
- from a subsistence harvest in preference to a commercial, exploratory, or scientific harvest, or sport or trophy hunt; and
- for larger and more representative samples.

Where harvest data are available by sex and age-class (e.g. for large mammals), weights were determined for those categories and applied accordingly.

Where more than one data set meets the above criteria, the mean value of all of them, unweighted for sample size, has been used as the standard value. Where there is notable weight variation among regional populations or stocks, values are recorded separately and applied to the community that harvests that population or stock.

Results

Edible weights for 10 species or species groups of mammals (including four subspecies of caribou), 12 species or species groups of birds, and 11 species or species groups of fish (including four separate Arctic char populations) are presented in Table 1. These weights are applied to the 10-year annual mean harvest of each species in order to provide an estimate, at the regional level, of available country food by species.

Total mean annual availability of country food (1998-1997) was approximately 332 457 kg, of which an estimated 41.3% was derived from large ungulates, 32.9% from fish, 17.2% from marine mammals, 6.0% from birds, and 2.5% from small mammals. Species for which values are provided are believed to account for approximately 99% of the regional harvest by weight. Using the 1991 population of 2890 Inuvialuit in the ISR (Statistics Canada 1994), mean annual per capita availability of country food was 115.0 kg.

Discussion and Conclusions

Edible weight values for most of the species utilized in the ISR have been developed through a standard method and set of criteria, and presented in a consistent and comprehensive format. A qualitative evaluation of the robustness of specific values has been provided (Usher 2000). Data users seeking to apply the values presented in this report to other parts of the Arctic may thus evaluate and select among original data sources, and conversion factors used to obtain edible weights. Where whole body weights given in the report (Usher 2000) fully meet the selection criteria noted above, they should be regarded as generally applicable. However, even where sample sizes are large (see criteria above) and data are precisely recorded, edible weights can be estimated only approximately at best, because of differences among harvesters (and between harvest circumstances) with respect to hunter selectivity, butchering practices, and food preferences. Data users seeking to apply the values presented in this report outside the ISR are therefore advised to take into account any local variations of which they are aware, when applying the edible weight values provided in this report.

Directions for future research include:

1. To ascertain or verify certain ambiguous or incomplete data through field projects, chiefly using local knowledge.
2. To analyze trends and variance in country food availability annually and seasonally at the community level, using recently available harvest survey data for the 10-year period.
3. To analyze these data in combination with recently obtained (but not yet available) diet survey data (Kuhnlein, this volume).

Project Completion Date

The project is complete and is in press, available from the Joint Secretariat, Box 2120, Inuvik, Northwest Territories, X0E 0T0.

Table 1. Mean annual country food yield by species, Inuvialuit Settlement Region, 1988-1997

Species	Mean Annual Harvest (kg)	Edible Weight per Animal (kg)	Total Food Weight (kg)
Marine Mammals			*57 320.00*
Ringed seal	1085	13.00	14 105.00
Beluga	129	335.00	43 215.00
Game Mammals			*137 259.00*
Caribou (Porcupine)	685	37.00	25 345.00
Caribou (Bluenose)	1868	36.00	67 248.00
Caribou (Dolphin/Union)	467	33.00	15 411.00
Caribou (Banks)	94	29.00	2726.00
Muskox	327	69.00	22 563.00
Moose	28	140.00	3920.00
Dall's sheep	2	23.00	46.00
Small Mammals			*8387.40*
Hare*	960	1.00	960.00
Muskrat	10 019	0.70	7013.30
Beaver	9	13.50	121.50
Lynx	77	3.80	292.60
Birds			*20 006.75*
Snow goose	5871	1.70	9980.70
White-fronted goose	1761	1.70	2993.70
Canada goose	658	1.05	690.90
Brant	489	1.00	489.00
Swan	127	4.75	603.25
Eider*	3281	1.30	4265.30
Oldsquaw	75	0.60	45.00
Mallard	225	0.85	191.25
Scoter*	167	0.90	150.30
Pintail	60	0.65	39.00
Wigeon	269	0.55	147.95
Ptarmigan	1026	0.40	410.40
Fish			*109 483.80*
Arctic char (Delta)	1170	0.65	760.50
Arctic char (Paulatuk)	2631	1.60	4209.60
Arctic char (Holman)	7847	1.55	12 162.85
Arctic char (Sachs Hbr)	600	0.70	420.00
Lake trout	5087	1.30	6613.10
Broad whitefish	23 184	1.65	38 253.60
Lake whitefish	8129	1.25	10 161.25
Whitefish spp**	1674	1.45	2427.30
Inconnu	5334	2.55	13 601.70
Arctic cisco	17 548	0.45	7896.60
Pacific herring	5905	0.20	1181.00
Herring/Cisco spp**	3197	0.30	959.10
Loche	3696	1.40	5174.40
Pike	2574	2.20	5662.80
Total			**332 456.95**

* Hare, eider and scoter are species groups, reported non-specifically to the Inuvialuit Harvest Study.

** Species groups, reported non-specifically to the Inuvialuit Harvest Study.

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Inuvik Regional Human Contaminants Monitoring Program

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Abstract

The objective of the study is to establish baseline levels of specific heavy metal and organochlorine contaminants in the blood of women and their newborns, from communities in the Inuvik Region, Northwest Territories. The study examines traditional/country food consumption as a possible contaminant exposure pathway, while measuring organochlorines and metals in maternal and cord blood, as well as mercury levels in maternal hair. The focus of the 1999-2000 fiscal year was to complete sample collection, consolidate laboratory results and conduct statistical analysis. Collection of data from this region completes the Northwest Territories/Nunavut database on maternal and cord blood. One hundred and four women from Inuvialuit, Gwich'in and non-native backgrounds participated in the study. The results are similar to existing data from other regions in the Canadian North and confirm the importance of traditional/country foods among women of reproductive age. Levels of exposure to contaminants were generally within guideline levels, with only a few exceptions. Communication of results to the communities will continue into the 2000-2001 fiscal year.

Objectives

1. To obtain values for the concentrations of organochlorine and metal contaminants in maternal and umbilical cord blood samples, and the concentration of mercury in hair samples from pregnant women in the Inuvik region.
2. To assess exposure to these contaminants through the frequency of traditional/country food intake and certain other lifestyle factors.
3. To describe any relationship between contaminant levels in blood and hair samples and frequency of consumption of traditional/country foods and selected lifestyle factors.

4. To derive relationships between dietary methylmercury (MeHg) exposure estimates obtained from predictive dietary exposure models and from biomarker-based methods.

Introduction

Low levels of environmental contaminants, such as organochlorines and heavy metals, are found in the northern ecosystem, arriving mainly through long-range atmospheric transport. These persistent contaminants have been detected throughout arctic ecosystems including air, surface seawater, snow, fish, marine mammals, sea birds, and terrestrial plants and animals (Jensen et al. 1997). A source of possible contaminant uptake in humans is through food consumption, since contaminants bioaccumulate and biomagnify in the food chain.

Many factors influence the amount of human exposure to contaminants through foods. This includes the concentration of contaminants in food, the size of each serving, and the frequency in which foods are consumed. In this study, the frequency in which participants consume traditional/country foods was examined. Past contaminant and dietary studies have detailed the concentration of contaminants in foods and relative portion sizes for people in the Inuvik Region (Receveur et al. 1996). Factors other than diet may influence the levels of contaminants measured in blood and hair of people. These include the use of tobacco products and occupational exposure. These factors may help to account for any unusual findings.

Increasing concern about long-range transport of contaminants prompted studies to establish a baseline of contaminant levels existing in Northerners, particularly vulnerable populations such as mothers and newborns. The results from the Inuvik region will complete the Northwest Territories (NWT) and Nunavut dataset on contaminants in maternal and cord blood. This assessment was designed in 1997-1998, based on the model used by the Mackenzie/Kitikmeot Cord Blood Monitoring Program (Rohlmann 1996).

Activities in 1999-2000

The focus of the 1999-2000 fiscal year was to complete the sampling portion of the study, consolidate data and conduct statistical analysis. Communication and community involvement continued to be important factors in the delivery of the program.

Collection of samples (hair and blood), and interviews with participants continued until the end of June 1999. Between April and June 1999, 15 individuals were recruited to participate in the study. The total number of individuals participating in the study was 104 pregnant women.

Levels of PCBs (polychlorinated biphenyls), organochlorines, toxaphene and fatty acids were determined from blood samples through analysis at the Centre de Toxicologie de l'Université de Laval at Laval University Hospital Centre. The MeHg testing in hair was completed by Health Canada (Indian and Northern Health Laboratory, Medical Services Branch). All results were forwarded directly to the program coordinator's office and entered into a database. Blood concentrations of contaminants and MeHg in hair were summarized for the region using descriptive statistics.

A preliminary report was produced, summarizing the frequency of consumption of traditional/country foods, and the health and lifestyle characteristics of the participants, based on responses to a questionnaire. Results were analyzed using descriptive statistics for traditional/country food consumption, demographics and lifestyle characteristics.

Dietary data were forwarded to the Centre for Indigenous People's Nutrition and Environment (CINE), for further statistical analyses. The analyses included quantifying traditional/country food consumption and identification of dietary patterns related to contaminant exposure.

CINE is using these data to develop a model which can be used to identify those parameters that are suitable indicators of exposure to MeHg and that allow for relationships between these parameters. This model can then be used to interpret results generated from other similar monitoring programs. These results will be reported by CINE.

The following activities were also completed in the 1999-2000 fiscal year:

- As part of the ongoing communications, newsletters were issued to participants and organizations interested in the study.
- The coordinator was involved with a contaminants workshop that was held in the Gwich'in settlement region, organized by the contaminants coordinator.
- The coordinator took part in a health fair in Ft. McPherson, NWT, answering questions about environmental contaminants and discussing the monitoring program.
- The coordinator attended several meetings to discuss the study, including discussions with the Government of the Northwest Territories in Yellowknife, and a local presentation with community health representatives. Health workers in the region were provided with

continuing information through presentations at medical rounds in the Inuvik Hospital.

- The coordinator delivered a presentation at a local high school "career week" initiative. The role of research in northern contaminants was discussed.
- The coordinator attended a risk assessment workshop in Deline, NWT, hosted by the Department of Indian Affairs and Northern Development in the spring of 2000.
- The coordinator attended an elders/scientists retreat in Yellowknife, hosted by the Dene Nation. This retreat provided great insight into the needs of community members and their concerns, as well as providing an education in traditional knowledge.
- A NWT Scientific Research License was issued for activities conducted in 2000.

Results

There were 104 participants in the study. Of these, 102 completed questionnaires and 73 provided hair samples for analysis. A total of 185 blood samples (95 maternal and 90 cord) were collected.

Demographic characteristics

The ages of the participants ranged from 15 to 45 years, with a mean age of 25.9 years (± 6.1 years). Participants identified themselves in one of three ethnic classifications; 44% identified themselves as Gwich'in, 33% as Inuvialuit and 23% as non-native. More than 90% of the respondents considered themselves to be low or medium consumers of traditional/country foods, relative to others in their community.

Blood

The blood samples were analyzed for a suite of substances such as: organochlorines, including PCBs (total as Aroclor 1260), congeners 28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183 and 187, aldrin, dieldrin, α-chlordane, γ-chlordane, β-HCH (β-hexachlorocyclohexane), oxyxchlordane, *cis*-nonachlor, *trans*nonachlor, *pp'*DDE, *pp'*DDT, heptachlorepoxide, HCB (hexachlorobenzene), mirex, and toxaphene; heavy metals, including cadmium, lead, and mercury (Hg) (total and inorganic); trace metals, including copper, selenium and zinc; and lipids.

The geometric mean and range of maternal and cord blood results for the four contaminants that have human health guidelines are presented in Table 1. The guidelines are consistent with those used in previous blood monitoring programs (Moss 1997). For calculations, non-detected values were replaced by a value equal to half of the limit of detection for the respective contaminant.

Mercury in hair

Hair grows at approximately 1 cm per month. By studying Hg in hair, we have a temporal view of exposure to Hg going back as many months as the hair was centimetres long. It provides an estimate of the highest exposure during preceding months, and also the length of any significant exposure.

In 42 of the 73 hair samples collected, there was at least one segment of hair that had concentrations exceeding the detection limit (DL = 0.4 $\mu g \cdot g^{-1}$). The geometric mean and range of Hg in maternal hair, as well as Health Canada guidelines are presented in Table 2. Hair data are the average concentrations of those measured in up to 12 one-centimetre segments of hair from each of the participants. Concentrations less than the detection limit were assumed to be 0.2 $\mu g \cdot g^{-1}$.

Dietary exposure to contaminants

Estimates of yearly traditional/country food consumption ($kg \cdot person^{-1} \cdot year^{-1}$), obtained from the food frequency questionnaires, were multiplied by the expected

Table 1. Geometric mean and range of maternal and cord blood in the Inuvik region for contaminants with known blood guidelines from Health Canada

	Maternal Blood*		**Cord Blood****		**Guidelines**	
Contaminant	**Mean** $(\mu g \cdot L^{-1})$	**Range** $(\mu g \cdot L^{-1})$	**Mean** $(\mu g \cdot L^{-1})$	**Range** $(\mu g \cdot L^{-1})$	**Level of Concern** $(\mu g \cdot L^{-1})$	**Level of Action** $(\mu g \cdot L^{-1})$
Lead	22.73	5.0 - 112.0	16.66	5.0 - 135.0	N/A	100
Cadmium	0.76	0.1 - 7.1	0.11	0.1 - 0.9	5.0	N/A
Mercury (total)	1.2	0.1 - 24.3	1.61	0.1 - 38.5	20.0	100
PCBs (Aroclor1260)	1.34	0.26 - 8.0	0.19	0.01 - 2.50	5.0	100

* n = 94 (lead, cadmium, mercury); n = 95 (PCBs)

** n = 89 (lead, cadmium, mercury); n = 85 (PCBs)

Table 2. Methylmercury levels in hair of women in the Inuvik region and guidelines from Health Canada

Contaminant	Maternal Hair		Guidelines	
	Mean ($\mu g{\cdot}L^{-1}$)	Range ($\mu g{\cdot}L^{-1}$)	Level of Concern ($\mu g{\cdot}L^{-1}$)	Level of Action ($\mu g{\cdot}L^{-1}$)
Methylmercury (n = 73)	0.20 - 7.90	0.56	6	30

contaminant concentration in each food item. Expected contaminant concentrations were obtained from the CINE database (Chan 1998) and reflect specific data obtained from the Inuvialuit and Gwich'in regions. Estimated exposure levels were converted to daily figures and appear reasonable when correlated with biological parameters. Table 3 indicates the number of records above intake guideline levels for each contaminant studied and the corresponding main food source of contamination.

Discussion and Conclusions

The region served by the Inuvik Regional Health and Social Services Board is the last area to complete the cord blood monitoring program in the NWT and Nunavut. Blood levels of contaminants from this region are low, and are similar to those of results from other communities in the Canadian North, in particular, the region served by Mackenzie Regional Health Services (Seddon et al. 1998). Levels of Hg and PCBs tended to be lower in this region compared to areas of the Eastern Arctic (Dewailly and Gilman 1998, Moss 1997). Indigenous peoples of the circumpolar region, and certainly within the Inuvik region itself, represent different ethnic, historical and cultural backgrounds. As well, climatic conditions and traditional diets vary. These differences will influence exposure levels to environmental contaminants.

The majority of values for contaminants in blood were within expected values. The Level of Concern established for contaminants with known guidelines was rarely exceeded. For maternal blood levels, only 3% of participants exceeded this level for lead, 4% for cadmium, 1% for total Hg and 5% for PCBs (as Aroclor 1260). For cord blood results, only 3% of participants exceeded the level of concern for lead and 2% for total Hg. Results of this study show that cord blood concentrations of Hg are 1.5 times higher than the maternal blood concentrations. None of the samples tested had amounts that exceeded those levels given for Level of Action.

MeHg has been measured in hair samples of Canadians by Health Canada for over 20 years. The highest levels have been found in Quebec, followed by Ontario and the NWT, with the exception of Inuit who consume sea mammals and fish, who have higher levels (Wheatley and Paradis 1996). Hair Hg levels from this study are low, with only 3% of participants exceeding the level of

Table 3. Number of food records exceeding contaminant intake guideline levels and main food contributors

Number of records above intake guideline levels* (total n = 102)	Main food contributors in records exceeding guideline levels	Food source intake ($kg{\cdot}person^{-1}{\cdot}year^{-1}$)	Exposure ($\mu g{\cdot}day^{-1}$)	Estimated exposure from food source (%)
Cadmium (n = 1)	Cloudberries	30	73.9	87
Lead (n = 8)	Caribou	59 ± 22	126.6 ± 47.9	38
	Grouse/ptarmigan	5.9 ± 11.3	113.7 ± 217.4	35
Mercury (n = 4)	Beluga meat	7.7 ± 13.7	44.5 ± 78.9	52
	Trout	8.1 ± 3.1	12.4 ± 3.1	15
Arsenic (n = 2)	Beluga fat	40.0 ± 14.1	140.2 ± 49.6	67
	Beluga meat	14.1 ± 20.0	50.3 ± 71.1	24
Chlordanes (n = 24)	Beluga fat	5.9 ± 11.2	32.3 ± 61.2	96
HCB (n = 3)	Beluga fat	30.0 ± 20.0	54.9 ± 36.6	98
PCBs (n = 6)	Beluga fat	18.2 ± 18.1	181.2 ± 180.2	94
Toxaphene (n = 23)	Beluga fat	6.1 ± 11.4	102.9 ± 190.5	95
Dieldrin (n = 12)	Beluga fat	9.9 ± 15.1	21.8 ± 33.2	86
HCHs (n = 2)	Beluga fat	40.0 ± 14.1	46.3 ± 16.4	97

* Guideline levels ([Provisional] Tolerable Daily Intakes) were calculated for a person of 60 kg using the following criteria ($\mu g{\cdot}day^{-1}$): cadmium = 1, lead = 3.57, mercury = 0.71, arsenic = 2, chlordanes = 0.05, HCB = 0.27, PCBs = 1, toxaphenes = 0.2, dieldrin = 0.1, HCHs = 0.3.

concern. These results are consistent with past findings of Hg levels in this region (Wheatley and Wheatley 1988).

The average frequency of consumption of many species of traditional/country foods by pregnant women in the Inuvik region can be described as moderate, however, a few women are frequent consumers of specific foods. Exposure levels were generally within guideline levels with only a few exceptions (Table 3). It is clear that any food, if consumed in large enough amounts, can be a significant source of contaminants (e.g. cloudberries in one record). Aside from this, some clear patterns emerge:

- Moderate amounts of caribou meat combined with small amounts of fowl meat can constitute a significant source of lead exposure.
- Small amounts of beluga meat and trout are the main sources of Hg in individuals exceeding guideline levels.
- Consuming beluga fat in excess of 5 to 6 kg per year puts the consumer above intake guideline levels, first for chlordane, then as the yearly consumption increases further, toxaphenes, dieldrin, PCBs, HCB and finally HCHs at levels of consumption reaching approximately 40 kg per year.

The Human Contaminants Monitoring Program will contribute to providing an overall picture of contaminant levels in humans throughout the north. Traditional/country foods are economically and culturally important and are highly valued by northern Aboriginal people. Northerners should continue to eat a variety of traditional/country foods for a balanced diet. The Inuvik Human Contaminants Monitoring Program will continue for a brief period of time into the 2000-2001 fiscal year to provide results and communications to the communities within the region. Participants will receive individual results for those contaminants with known guidelines, as well as general contaminant information. Any participant with levels above the Level of Concern will receive information on how to reduce future contaminant intake. This communications initiative will help interpret the results and aid Northerners in making healthy lifestyle choices.

Expected Date of Completion

June 30, 2000

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onitoring the Health of Peoples and Ecosystems and the Effectiveness of International Controls

iotic Monitoring and Modelling

New Persistent Chemicals in the Arctic Environment

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Abstract

This report updates the investigation of new chemicals in the Arctic environment, including brominated flame retardants, polychlorinated naphthalenes (PCNs), coplanar polychlorinated biphenyls, short-chain chloroparaffins (SCCPs), current use pesticides, pesticide enantiomers, chlorinated phenols and haloacetic acids. Methods were developed for brominated flame retardants other than the diphenyl ethers: tetrabromobisphenol-A (TBBPA), hexabromocyclododecane (HBCDD) and polybrominated biphenyls (PBBs). An initial examination of 12 Arctic air composites showed traces of TBBPA and PBBs in one to two samples, and undetectable levels in the rest. HBCDD was not found in any sample. Brominated diphenyl ethers (BDPEs) were determined in archived filter and PUF samples from Tagish, Yukon, collected in 1994-1995. High levels (100 to 800 pg·m^{-3}) of total BDPEs were detected during the summer months. Methods were optimized for gas chromatography-mass spectrometry analysis of 25 current use pesticides (including some metabolites). Seawater samples from Holman Island, Northwest Territories were analyzed but all compounds were below limits of detection except for endosulfan which was present at 1-3 pg·L^{-1}. PCNs in composite air samples from Alert and Dunai ranged from 0.01 to 2.3 pg·m^{-3}. Higher concentrations were found in late fall through early spring. This trend suggests an association between PCNs and combustion haze aerosols. A capillary gas chromatography method was developed which separates all the isomeric penta- and hexachloronaphthalenes. Concentrations of SCCPs in sediments collected on the 1997-1998 Surface Heat Budget of the Arctic and Joint Ocean Ice Studies projects ranged from 4.8 to 77.4 ng·g^{-1}. A clear decreasing trend was observed from south to north, and western Arctic concentrations were lower than in the eastern Arctic. SCCPs in sediments displayed a shorter carbon chain length and lower chlorinated C_{10} and C_{11} formula groups relative to the commercial PCA-60 formulation.

Heptachlor-exo-epoxide and *trans*-chlordane were non-racemic in seven composite air samples from Alert and Dunai. Non-racemic chlordane in air suggests that a portion of the chlordane now being transported to the Arctic comes from older sources (e.g. soil emissions).

Objectives

1. To determine coplanar polychlorinated biphenyls (PCBs), polychlorinated naphthalenes (PCNs), brominated diphenyl ethers (BDPEs), chlorophenolic compounds, chloroparaffins and modern agrochemicals in air from Arctic monitoring stations.
2. To search for other "new" chemicals in the Arctic environment, not currently monitored by the Northern Contaminants Program (NCP) but of potential concern based on known persistence, extent of usage and toxicology.

Introduction

The rationale for undertaking a study of new chemicals in the Arctic environment was presented in last year's report to the NCP (Bidleman et al. 1999). "New" chemicals are those which are currently used and/or not presently included in monitoring programs. Substances undergoing investigation in this project include BDPEs (flame retardants), PCNs and coplanar PCBs (both components of dielectric fluids and formed as combustion by-products), polychloroalkanes (or SCCPs), industrial compounds, chlorophenols (wood preservatives), modern agrochemicals, pesticide photoproducts and enantiomers, and halo-acetic acids (HAAs). The research proposed here directly relates to the NCP Blueprint for *Monitoring the Health of Arctic Peoples and Ecosystems and the Effectiveness of International Controls*, Section 1.3, "Other Chemicals of Potential Concern"(Northern Contaminants Program 1999). Certain aspects also contribute to other sections of the Blueprint. Investigation of enantiomer ratios for chlordanes, α-HCH and toxaphenes will aid in detecting and interpreting trends in relation to emission control strategies by providing a means of distinguishing "recycled" pesticides from new releases.

Activities in 1999-2000

Sample acquisition

Surface sediment samples collected during the 1997 and 1998 Joint Ocean Ice Studies (JOIS) cruise track through the Canadian Archipelago to the Surface Heat Budget of the Arctic (SHEBA) site in the Canadian Basin interior were analyzed for SCCPs. Archived air samples from Arctic air monitoring stations, 1994-1995, were analyzed for PCNs, brominated flame retardants and BDPEs as well as pesticide enantiomers. Seawater samples were collected at Holman Island, Northwest Territories (NWT), using an Infiltrex in situ sampler with collection on XAD-2 resin.

Analytical methods development

Brominated flame retardants

Methods were developed for the non-BDPE flame retardants, which included hexabromocyclododecane (HBCDD), tetrabromo-bisphenol A (TBBPA) and poly-brominated biphenyls (PBBs). HBCDD and PBBs were run on a 30 m HP-5 column, 250 μm i.d. x 0.25 μm film thickness using He at a flow rate of 1.2 mL·min^{-1}. GC program: initial temperature 80°C for 1 min followed by a ramp of 15°C/min to 200°C and held for 1 min. This was followed by a second ramp to 250°C and held for 5 min, and then followed by a third ramp of 5°C/min to 300° and held for 15 min. The injection temperature was 270°C.

Detection was by high resolution mass spectrometry, and the method acquisition consisted of five functions ranging from lock mass 230.98563 (F1) to 430.97285 (F5). F1 = mono (M+), F2 = di (M^+) and tri (M^+), F3 = tri (M^+) and tetra ($M-Br^+$), F4 = tetra ($M-Br^+$) and penta ($M-2Br^+$), F5 = hexa ($M-2Br^+$).

TBBPA was derivatized in air sample extracts using 100 μL of 1:1 acetic anhydride/pyridine for 30 min at 60°C, followed by a 1 mL wash with deionised water prior to analysis. The derivatized TBBPA was run on the same 30 m HP-5 column as above using the GC program: initial temperature 80°C for 1 min followed by a ramp of 20°C/min to 200°C and held for 1 min. This was followed by a second ramp of 10°C/min to 300°C and held for 13 min. The injection temperature was 270°C. Method acquisition consisted of one function, monitoring $M-Br^+$ with a lock mass of 492.9697.

Current use pesticides

GC-MS conditions for analysis of a suite of current use pesticides (see Table 1 for list) were optimized for electron ionization (EI), positive chemical ionization (PCI) and electron capture negative ionization (ECNI) conditions on a Hewlett Packard 5973 MSD. Pesticides were separated using a 30 m × 0.25 mm i.d. DB-5 column with helium carrier gas. Limits of detection were calculated based on peak signal-to-noise ratios of 3 × noise level using a series of calibration standards having concentrations of 0.500 ng·μL and higher.

Polychlorinated naphthalenes

Several PCN congeners coelute on the DB-5 column used for routine analysis. We investigated other columns and found that one cyclodextrin-based column, Rt-βDEXcst (Restek Corp.), was able to resolve all the isomeric penta- and hexa-CNs (Helm et al. 1999). This is remarkable, as other researchers have tried many different GC columns and failed to achieve this degree of separation. Some penta- and hexa-CNs elicit dioxin-like effects and have been assigned toxic equivalency factors (TEFs). The coeluting penta-CNs 52/60 and hexa-CNs 66/67 are especially bioaccumulative and are the most abundant members of their homologue groups in Arctic air. The TEF for CN-66 is nearly four times higher than for CN-67 (Blankenship et al. 2000). Until now these CNs could not be separated by capillary GC, but they are resolved on the RtβDEXcst column. This column will also allow for the use of isomer ratio analysis for congeners which typically coelute and have the same vapour pressure (Lei et al. 1999), as well as the use of marker congeners from combustion, to aid in identifying sources which contribute to the PCNs observed in air.

Results and Discussion

SCCPs in sediments

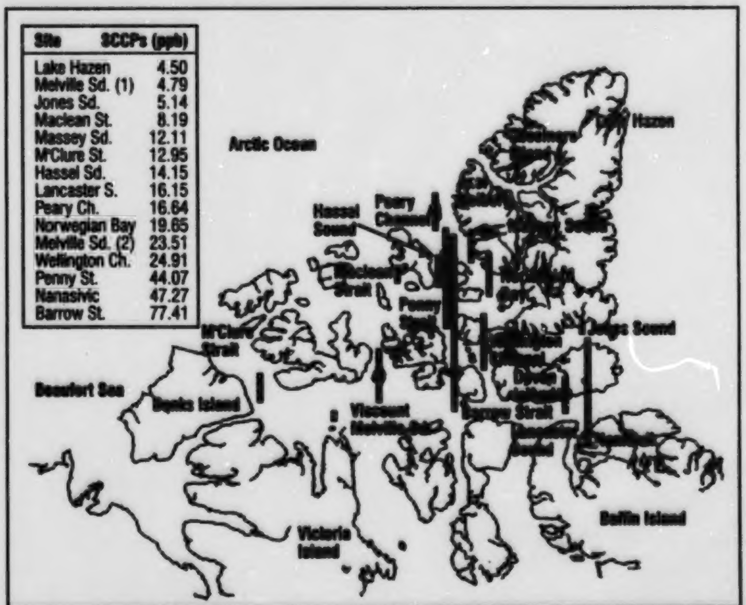
Concentrations of SCCPs in sediment ranged from 4.8 to 77.4 ng·g^{-1}. A clear decreasing trend was observed from south (Barrow Strait) to north (Peary Channel), and western Arctic (M'Clure Strait/Viscount Melville) sediment concentrations were lower than in the eastern Arctic (Nanisivik/Lancaster Sound) sediments (Figure 1). Tomy et al. (1999) reported a SCCP concentration of 4.5 ng·g^{-1} in the surface slice of a sediment core collected from Lake Hazen on Ellesmere Island. Relative to the commercial PCA-60 formulation (C_{10}-C_{13}, 60% Cl) the shorter carbon chain length and lower chlorinated C_{10} and C_{11} formula groups are clearly predominant (Figure 2). This result is consistent with the trend of increasing vapour pressures with decreasing carbon chain length and degree of chlorination reported by Drouillard et al. (1998) and their corresponding ability for long-range atmospheric transport. A trend of decreasing Henry's law constants with degree of chlorination was also reported, however no major differences were observed based on carbon.

PCNs in air

The ΣPCN concentrations in archived air samples from 1994-1995 ranged from 0.01 to 0.99 pg·m^{-3} at Alert, Nunavut, and from 0.03 to 2.33 pg·m^{-3} at Dunai Island, Russia, (Figure 3) and were comparable to levels reported for these sites in 1993 by Harner et al. (1998). ΣPCN concentrations at the high Arctic sites were lower than those observed at lower Arctic latitudes over the eastern Arctic Ocean, Norwegian Sea and Barents Sea (Harner et al. 1998). Analysis of coplanar PCBs in these samples has not yet been completed.

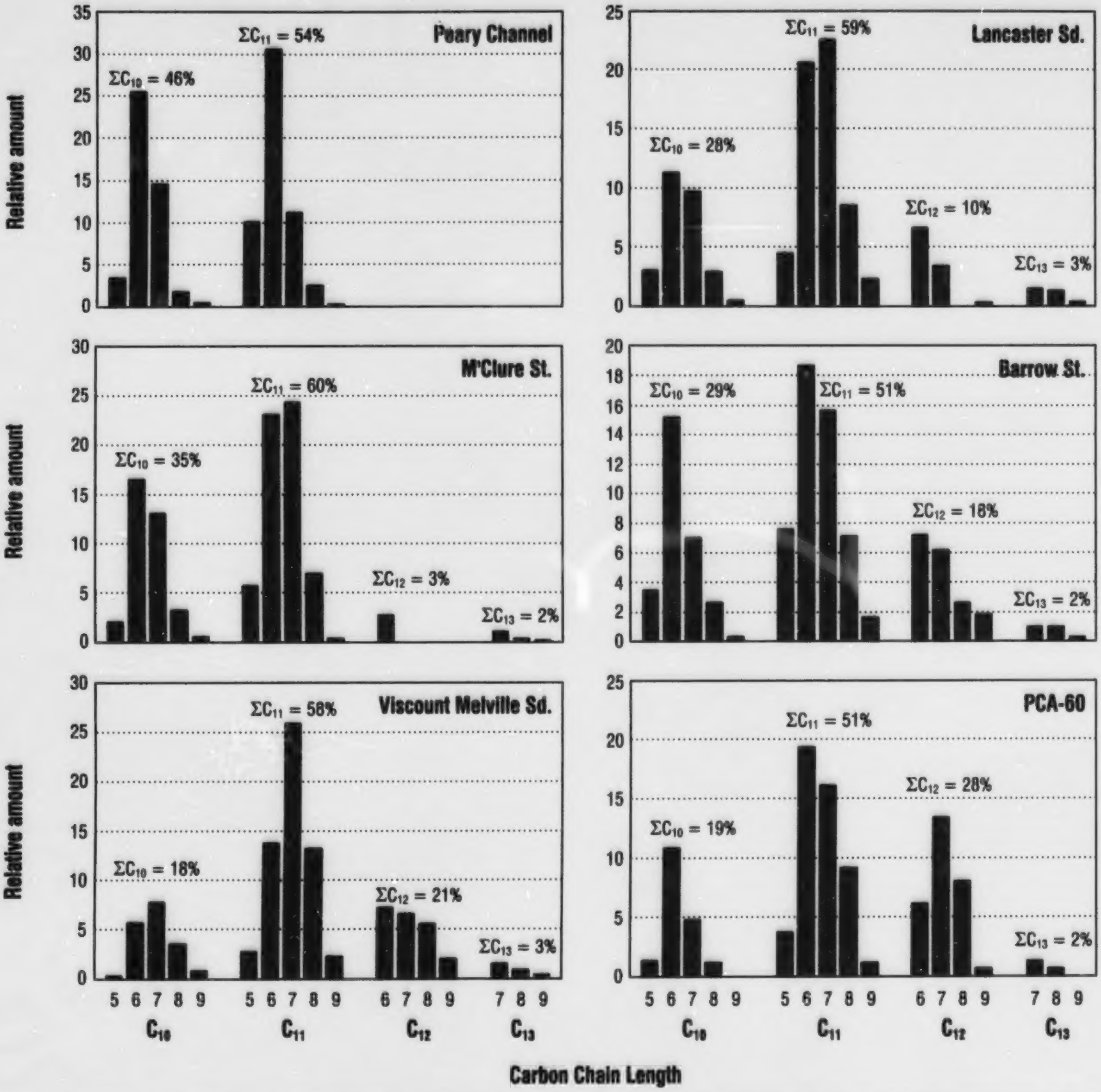
Seasonal variations in ΣPCNs were observed at both the Alert and Dunai sites, with higher ΣPCN concentrations in winter months (October to April) than in the summer

Figure 1. SCCP concentrations (ng·g^{-1}) in sediment samples from the Canadian Arctic



Site	SCCPs (ppb)
Lake Hazen	4.50
Melville Sd. (1)	4.79
Jones Sd.	5.14
Maclean St.	8.19
Massey Sd.	12.11
M'Clure St.	12.95
Hassel Sd.	14.15
Lancaster S.	16.15
Peary Ch.	16.64
Norwegian Bay	19.65
Melville Sd. (2)	23.51
Wellington Ch.	24.91
Penny St.	44.07
Nanasivic	47.27
Barrow St.	77.41

Figure 2. SCCP formula group profiles for surface sediment from the Canadian Archipelago and the PCA-60 commercial formulation (C_{10}-C_{13}, 60% Cl)

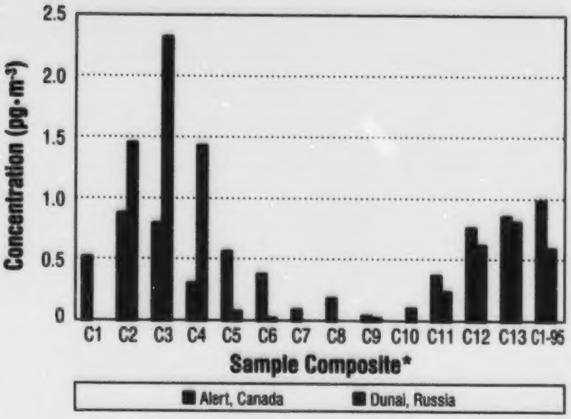


months (May to September) (Figure 3). This trend was found for not only the tri- and tetra-CN congeners which are the most abundant, but also for the more toxic and bioaccumulative penta- and hexa-CN congeners. This may be due to association with Arctic haze during winter and spring or effects of photolysis in summer. The trend may also be an artifact caused by breakthrough loss of tri- and tetra CNs during the warm months, although this would not be expected to occur for the less volatile CNs. Analysis of backup PUFs in the coming year will allow this to be evaluated.

Brominated flame retardants in air

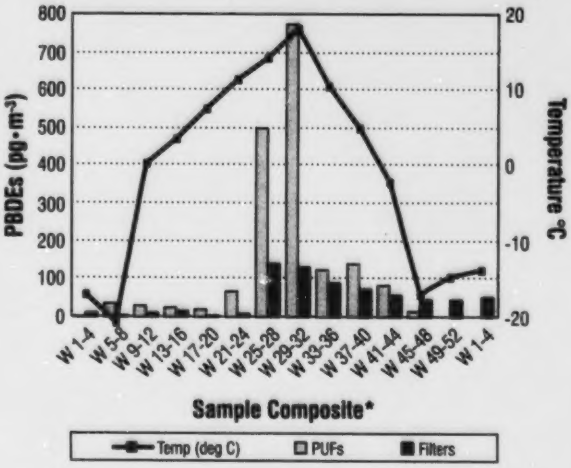
Four samples each from Alert, Tagish and Dunai were analyzed for brominated flame retardants other than BDPEs. The compounds sought were hexabromocyclododecane (HBCDD), tetrabromo-bisphenol A (TBBPA) and polybrominated biphenyls (PBBs). Preliminary results are listed in Table 1. One filter sample from Alert (summer) and one from Tagish (winter) appeared to contain some PBB congeners, but levels were very low and further concentration of the subsamples will be required for further analysis. No HBCDD was detected in any of the

Figure 3. ΣPCNs in air at Alert, Canada, and Dunai, Russia, 1994-1995



* Composite sample numbers on the x-axis correspond roughly to months (13 composites per 12 month period).

Figure 4. Total mono- to heptabromodiphenyl ethers in air (PUFs and filters) in archived air sample extracts, Tagish, Yukon, 1994-1995 (pg·m⁻³)



* 4-week composite samples

samples analyzed. TBBPA was detected on the filter of one spring sample from Dunai at a concentration of 70 pg·m⁻³. Decabromodiphenyl ether was also not present on filters at detection limits of about 1 pg·m⁻³.

BDPEs were determined in archived air sample extracts from Tagish, Yukon. These samples represented composites of 4 weekly samples (i.e. weeks 1-4, 4-8, etc.) from January to December 1994 and January 1995. The BDPEs were determined by high resolution MS as described previously (Bidleman et al. 1999). Relatively high levels of mono- to heptabromoDPEs were detected at Tagish with maximum concentrations occurring during the summer months (Figure 4). The much higher air temperatures at Tagish compared to Alert and Dunai combined with closer proximity to populated areas in southern Yukon may be the reason for the high levels although local sources cannot be ruled out. Field blanks showed only very low levels of the BDPEs. BDPE congeners 47 (2,4,2',4'-tetra) and 99 (2,4,2',4',5'-penta) were the major congeners in both PUFs and filters. A higher percentage of BDPE99 was on filters (8% to100%) compared to BDPE47 (2% to 26%), especially during the winter months.

Table 1. Preliminary results of analysis of Arctic air sample filters* for several brominated flame retardants including decabromodiphenyl ether

Compound	Alert pg·m⁻³	Tagish pg·m⁻³	Dunai pg·m⁻³
Hexabromocyclododecane (HBCDD)	< 1.8	< 1.8	< 1.8
Tetrabromo-bisphenol A (TBBPA)	< 0.07	< 0.07	< 0.07 - 70
Polybrominated biphenyls (PBBs)	< 0.03 - ~ 0.1	< 0.03 - ~ 0.1	< 0.03
Decabromobiphenyl ether	< 0.1	< 0.1	< 0.1

* Four extracts representing 4-week composites of filters were analyzed from each location

Pesticide enantiomers in air

Five of the monthly composite air samples from Alert and two from Dunai were analyzed for enantiomers of α-HCH, heptachlor-exo-epoxide (HEPX) and chlordanes. The α-HCH at these land-based stations was racemic, with an average enantiomer ratio, ER = (+)/(-) = 1.01 ± 0.015. Over ice-free areas of the Arctic Ocean, α-HCH is non-racemic due to sea-to-air exchange of the non-racemic α-HCH in surface water (Harner et al. 1999, Jantunen and Bidleman 1996).

The HEPX in all air samples was distinctly non-racemic; the average ER = 2.27 ± 0.69, with all but one sample lying between 1.87 and 2.12 and the other value = 3.83. There are two sources of HEPX to the atmosphere. One is photolysis of the pesticide heptachlor, which yields mainly racemic photoheptachlor and a smaller proportion of racemic HEPX (Buser and Müller 1993). The other is formation of HEPX from heptachlor in soils, which occurs with a strong preference for the (+) enantiomer (Aigner et al. 1998, Bidleman et al. 1998). Photoheptachlor has been identified in seal blubber and human milk samples from Arctic Canada (Zhu et al. 1995). However the non-racemic HEPX in air indicates that it is probably derived mainly from soil emissions rather than photolysis of heptachlor (Bidleman et al. 1998).

The HEPX in seawater collected on the AOS-94 expedition had an ER = 1.61 ± 0.08 (Jantunen and Bidleman 1998). In the Arctic food chain, the ERs of HEPX are 1.2-1.4 in Arctic cod, 0.7 in ringed seal blubber, 2.1 in polar bear fat and 3.3 in polar bear liver (Wiberg et al. 2000). Thus HEPX is atmospherically delivered to the Arctic Ocean in non-racemic form and is then further degraded or selectively bioaccumulated in the food chain.

Chlordane ERs for the seven air samples averaged 0.91 ± 0.039 for *trans*-chlordane (TC) and 0.98 ± 0.067 for *cis*-chlordane (CC). ERs for the chlordane standards (6 replicate injections) were 0.98 ± 0.028 (TC) and 1.01 ± 0.049 (CC). The ER of TC in air is significantly lower ($p < 0.01$) than for the standard. Ratios of TC/CC in Arctic air have also declined from the mid-1980s to the 1990s (Bidleman 1999). These observations suggest that the Arctic is now receiving some chlordane that has been "recycled" from old sources, possibly agricultural soils where non-racemic chlordanes have been found (Aigner et al. 1998). Non-racemic chlordanes have also been found in air of the Great Lakes region (Bidleman and Falconer 1999, Ulrich and Hites 1998). Racemic chlordanes have been reported in air of the southern United States (Bidleman and Falconer 1999, Jantunen et al. 2000) and in Norway (Buser and Müller 1993).

Current use pesticides in seawater

The optimization of GC-MS conditions for current use pesticides showed that EI was the best mode for general purpose since all organophosphates, organonitrogens and triazines had lowest limits of detection (LODs) in this mode. ECNI may be better for selected compounds such as dacthal, because it is more selective and background is suppressed. PCI had the most predictable fragmentation pattern but also the highest LODs in general. The LODs for a 100 L seawater sample are given in Table 2 for the EI and ECNI modes.

Three 100L seawater samples collected at Holman Island, NWT using XAD-2 columns in May 1999 were analyzed for the suite of current use pesticides using both EI and ECNI MS. The XAD extracts were chromatographed on silica columns to separate PCBs from most organochlorines. The only current use pesticide detectable was endosulfan which was present at 1 to 4 $pg \cdot L^{-1}$. This widely used insecticide has been found previously in Arctic ocean waters. Many of the compounds listed may not have fully eluted from the silica cleanup column with dichloromethane:hexane (1:1). Further work is underway to optimize the cleanup steps for the seawater extracts so that all the pesticides are efficiently recovered.

Expected Project Completion Date

March 31, 2001

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Table 2. Survey of current use pesticides and degradation products in extracts of seawater*

Pesticide (common name)	Class	EI-MS pg·L^{-1}	ECNI-MS pg·L^{-1}
alachlor	phenylamide	< 1	not determined
ametryn	triazine	< 3	< 224
atrazine	triazine	< 2	< 84
butralin	dinitroaniline	< 4	< 0.6
chloropyrifos	OP**	< 17	< 1
chlorothalonil	benzenedicarbonitrile	< 2	< 2
dachtal	benzenedicarboxylic acid	< 2	< 0.01
des-ethyl atrazine	triazine	< 3	< 154
diazinon	OP	< 3	< 1
diazinon-oxon	OP metabolite	< 2	< 172
dimethoate	OP	< 18	< 7
disulfoton	OP	< 2	< 6
endosulfan ($\alpha+\beta$)	chlorinated cyclodiene	1-4	36528
ethion	OP	< 1	< 0.6
fenitrothion	OP	< 11	< 0.3
flutriafol	triazole-ethanol	< 65	< 71
fonofos	OP	< 1	< 0.01
guthion	OP	< 16	< 3
hexazinon	triazinone	< 5	not determined
imidan	OP	< 3	< 0.9
methyl parathion	OP	< 50	< 1
metolachlor	phenylamide	< 2	< 229
pendimethalin	dinitroaniline	< 4	< 0.2
phorate	OP	< 4	< 3
primiphosmethyl	OP	< 2	< 48
terbufos	OP	< 1	< 10*

* All compounds except endosulfan were present at < LOD using either GC-EI-MS or GC-ECNI-MS.

** Organophosphate pesticide

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Northern Contaminants Air Monitoring and Interpretation

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Abstract

Atmospheric measurements of organochlorines and polycyclic aromatic hydrocarbons have been conducted at Alert, Northwest Territories since 1992. In this study, levels of atmospheric organochlorines from 1993 to 1997 have been examined. Air-surface equilibrium analyses confirmed previous results showing that fresh inputs of current-use pesticides reach the Arctic environment. Compound-specific seasonality and slightly decreasing atmospheric trends for most banned or restricted organochlorines were observed.

Objectives

1. To measure and understand the occurrence and trends of selected organochlorines (OCs) and polycyclic aromatic hydrocarbons (PAHs) in the Arctic atmosphere to determine whether concentrations are changing in response to national and international initiatives.
2. To provide insight into contaminant pathways (sources, transport, transformation and removal processes) to the Arctic environment.
3. To enable validation of models of toxic chemicals in the Arctic environment with atmospheric observations.
4. To operate a major long term trends measurement station at Alert, Northwest Territories, (in operation since 1992) in parallel with a western Russian Arctic station to contribute to the next assessments by the Northern Contaminants Program (NCP) and the Arctic Monitoring and Assessment Programme, and to advise Canadian negotiators in preparing contaminant control strategies.

Introduction

Atmospheric measurements of persistent organic pollutants, including herbicides, pesticides, synthetic industrial compounds and PAHs, have been made on a weekly basis in the Canadian (Alert, Tagish, Kingait [Cape Dorset]) and Russian Arctic (Dunai, Amderma) since January 1992. These measurements are essential since there has been growing concern about contamination of the Arctic by OCs and PAHs, whose environmental persistence and fate are influenced by a diversity of physical-chemical properties. Long-term time series will help to determine whether the atmospheric concentrations and deposition of priority pollutants in the Arctic is increasing or decreasing in response to various national and international control initiatives.

Activities in 1999-2000

Regular weekly atmospheric measurements of OCs and PAHs continued at Alert. This involved the collection, extraction and analysis of air samples. Similar measurements were conducted at Amderma, Russia.

Data analysis has been progressing with the recent publication of a paper describing episodes of rapid transport of contaminants to Tagish, Yukon (Bailey et al. 1999). A new post-doctoral fellow (H. Hung) has started to investigate the wealth of information contained in this growing database. The results and discussion sections below contain part of Dr. Hung's work that will be presented at the DIOXIN 2000 conference later this year. Specifically, the temporal patterns (1993-1997) of atmospheric OC levels at one of the stations, Alert, are examined.

Results

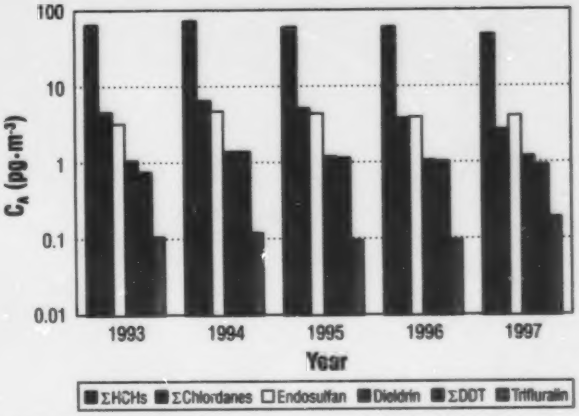
Mean annual concentrations (filter + polyurethane foam plugs) of selected OCs are given in Figure 1. α-HCHs (hexachlorocyclohexanes) and γ-HCHs, being the lightest of all the selected OCs, were the highest in concentration for all five years. β-HCH, heptachlor, *c*-nonachlor, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDE and trifluralin were detected at very low levels. The atmospheric concentrations were greatest in he following order during the first three years: ΣHCHs > Σchlordanes > endosulfan> dieldrin> ΣDDTs > trifluralin. In 1996 and 1997, endosulfan concentrations were higher than that of Σchlordanes.

Discussion and Conclusions

Levels of different organochlorines

The average concentrations of endosulfan did not change significantly during the five years of sampling, whereas that of Σchlordanes apparently decreased during 1994-1997 (Figure 1). Although chlordane has been deregistered in most European countries and North America since the 1980s (Barrie et al. 1992), its sole producer, Velsicol, has only permanently ceased production since May 1997 (Pesticide Action Network North America 1997). United States custom records have shown that between 1991 and 1994, at least 2028 metric tonnes of chlordane and 2584 metric tonnes of heptachlor were exported (Pesticide Action Network North America 1997). A decrease in emissions of these compounds has not been reflected in the Arctic atmospheric levels until recently. Dieldrin, an insecticide and the epoxidation product of aldrin, did not show much change in atmospheric concentration in the Arctic, although both aldrin and dieldrin were banned in Europe and North America before 1990, and world production ceased in 1991 (Siedenburg 1991).

Figure 1. Average annual concentrations of OC pesticides at Alert



Air-surface equilibrium

If equilibrium has been reached between the air and Arctic surfaces, the relationship between temperature, T (K), and air concentration, represented by partial pressure, P (Pa), can be expressed according to the Clausius-Clapeyron equation:

$$\ln P = m/T + b \qquad (1)$$

where m and b are compound-specific constants. m is, in fact, $-\Delta H_{SA}/R$, where ΔH_{SA} is the enthalpy of phase change between surface and air and R is the universal gas constant (Cortes et al. 1998). Regression results for the various OCs are given in Table 1. Significant correlation was observed for most of the compounds ($p < 0.01$). The slopes obtained for all compounds were lower than those calculated using only 1992-1994 data and were considerably lower than those obtained in temperate regions (Halsall et al. 1998). The conclusions drawn are generally the same as those described by Halsall et al. (1998). In summary, major current-use pesticides, such as the HCHs, DDTs, endosulfan and trifluralin have lower correlation with temperature due to fresh atmospheric inputs. Metabolites, such as *c*-chlordane, oxychlordane and heptachlor epoxide, showed higher correlation with temperature ($p < 0.001$) indicating revolatilization from Arctic surfaces.

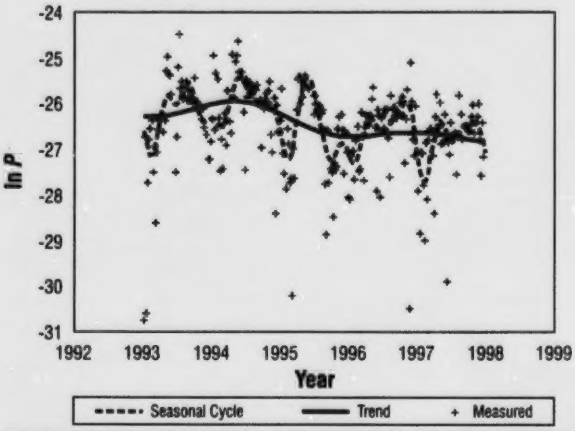
Table 1. Regression results of $\ln P$ (Pa) versus $1/T$ (1/K)

OCs	m	b	r^2	ΔH_{SA} kJ/mol
α-HCH	**227**	**– 22.7**	**4.6×10^{-3}**	**– 1.9**
γ-HCH	– 27	– 23.6	4.5×10^{-5}	0.22
dieldrin	– 2943	– 14.7	0.30	24
t-chlordane	– 854	– 23.9	0.04	7
c-chlordane	– 1930	– 19.0	0.17	16
t-nonachlor	– 2870	– 15.4	0.35	24
c-nonachlor	– 3919	– 13.6	0.54	33
oxychlordane	– 2195	– 18.1	0.24	18
heptachlor	**592**	**– 31.7**	**0.03**	**– 4.9**
heptachlor epoxide	– 3107	– 14.2	0.31	26
p,p'-DDE	1594	– 33.6	0.10	– 13
p,p'-DDD	– 600	– 26.8	0.03	5.0
p,p'-DDT	– 1055	– 24.2	0.06	88
endosulfan	1102	– 15.7	0.08	– 9.2
trifluralin	– 507	– 26.4	0.08	4.2

Underline indicates that slopes are not significant at the $p = 0.01$ level.

Bold type indicates compounds that were negatively correlated with temperature.

Figure 2. $\ln P$ of *c*-chlordane measured from January 1993 to December 1997 at Alert



Seasonality and time-series analysis

Seasonality of atmospheric levels was apparent for many of the OCs analyzed. The seasonal cycle and trend lines (Figure 2 for *c*-chlordane) were determined using a digital filter technique (Nakazawa et al. 1997). Both *t*- and *c*-chlordanes peaked around May to June and again around November to December. For both α- and γ-HCHs, peaks were observed around February and August during all five years. Elevated concentrations around February probably resulted from spring spraying of technical HCHs and/or lindane in Asia. Back trajectory analysis has indicated that in February, Alert was strongly influenced by air from Eurasia (Halsall et al. 1997). Although most of the DDT-related compounds were observed at very low levels, *p,p*'-DDE, the most abundant of all DDT-related compounds in air, showed distinct seasonal peaks. *p,p*'-DDE level usually peaked from October to December, then decreased slightly and peaked again around March to May, followed by a minimum during summer. In India, elevated levels of *p,p*'-DDE have been recorded during the dry season of January to May, probably due to oxidative dehydrochlorination of DDT to DDE in dry paddy soil (Ramesh et al. 1989). This may account for the elevated level of *p,p*'-DDE in spring at Alert. Not all the OCs showed concentration peaks during the warmest months. As mentioned above, levels of *p,p*'-DDE, *t*- and *c*-chlordanes soared during winter (about October to December). Cyclonic movement of air flow in winter causes air originating from the strong Siberian anticyclone to curve eastwards around the Arctic region (Barrie et al. 1992). This makes the dissipation of pollutants more difficult in winter. Besides, slow degradation rates at low temperatures and the lack of photodegration, due to the low levels of solar radiation, may result in elevated levels of certain compounds, such as *t*- and *c*-chlordanes, in winter.

Table 2 summarizes regression results of the calculated trends. Most of the compounds showed decreasing trends from 1993 to 1997 with statistically significant slopes ($p < 0.01$). γ-HCH was declining at a faster rate than α-HCH. This might be caused by photo-transformation of γ- to α-HCH during transport or past/present use of technical HCH, in which approximately 70% is α-HCH (Halsall et al. 1998). Although dieldrin showed a positive trend (despite its low r²), close examination of its seasonality showed that the amplitudes of the seasonal cycle decreased by about 50% from 1993 to 1997. The average growth rate had decreased from 0.2 pg·m^{-3} per year in 1993 to 0.08 pg·m^{-3} per year in 1997, with negative average growth rates in 1995 and 1996. Thus, from this analysis, a significant decline in dieldrin atmospheric concentrations is likely within the next few years despite its almost constant level from 1993 to 1997. All chlordane-related compounds showed significant decreasing trends, except heptachlor. This is probably an artifact of its extremely low concentrations in the Arctic. However, its degradation product, heptachlor epoxide, showed a significant negative slope as expected, since heptachlor has been restricted or banned in most countries during the 1970s and 1980s. Endosulfan, as a current-use pesticide, showed an increase in concentrations from

Table 2. Linear regression of calculated OC trends, on ln*P* (Pa) versus time series (year) plots, using digital filter technique

	Trends*	
OCs	**Slope**	r^2
α-HCH	–0.0410	0.79
γ-HCH	–0.142	0.88
dieldrin	0.0134	0.021
t-chlordane	–0.0834	0.48
c-chlordane	–0.169	0.84
t-nonachlor	–0.112	0.71
c-nonachlor	–0.0878	0.28
oxychlordane	–0.200	0.84
heptachlor	0.0899	0.10
heptachlor epoxide	–0.110	0.72
p,p'-DDE	–0.0366	0.054
p,p'-DDD	0.0385	0.54
p,p'-DDT	0.107	0.41
endosulfan	0.134	0.25
trifluralin	–0.0925	0.36

* Linear regression of trend calculated by digital filter technique.

early to mid-1993. Then, an almost steady level was maintained through the end of 1997.

Some of the OCs manifested similar trends in a study on male Pangnirtung beluga blubber (Stern, personal communication). In some cases, the trends may even be magnified in mammals since most of these compounds are hydrophobic. For instance, endosulfan concentrations in beluga blubber increased by a factor of 2.1 from 1982 to 1997 in the eastern Arctic.

Four years of data collected at Alert have demonstrated compound-specific seasonality and slightly decreasing atmospheric trends for most banned or restricted OCs. However, some pesticides still in use (e.g. endosulfan) do not present this behavior. Atmospheric measurements at Alert and Kingait will continue this fiscal year to provide additional data needed for a better understanding of the long-term trends of these chemicals. Data analyses will continue with an emphasis on chlordanes, endosulfan, DDT, DDE and HCHs to assess the trends, emission sources and transport pathways of these chemicals in the Arctic.

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Toxaphene in the United States: Use, Emissions, Residues and Environmental Impact

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Abstract

Toxaphene was widely used in the United States between 1947 and 1986, with total usage of about 490 kt, more than 40% of the total global use. Inventories of gridded toxaphene usage, emissions and residues in agricultural soil in the United States with 1/6° latitude by 1/4° longitude resolution have been created. Total toxaphene emissions were 194 kt between 1947 and 2000. In 2000, almost 20 years after banning the use of toxaphene, there are still around 30 kt of toxaphene left in the agricultural soil; 360 t of these toxaphene residues in the agricultural soil will emit to the air in the same year. Atmospheric concentrations of toxaphene in the Canadian Arctic seem to be responding to reductions of toxaphene emissions from soil in the United States. This shows that toxaphene residues in American agricultural soil could be a major source of toxaphene in the Arctic air. A continuation in the trend in declining air concentrations in the Arctic is expected as this compound decreases in soil in the United States due to emission and degradation. However, as the air toxaphene concentration drops, the net direction of air-sea gas flux will be reversed since toxaphene concentrations in Arctic waters have remained relatively unchanged. Thus, the Arctic waters containing toxaphene will become a source to the atmosphere that will likely maintain detectable atmospheric concentrations for some time in the future.

Objectives

1. To create gridded toxaphene emission inventories worldwide, including the major user, the United States.
2. To study the linkage between the toxaphene emissions and the toxaphene concentration in the Arctic air.

Introduction

Toxaphene (camphechlor) was once one of the most heavily used pesticides worldwide, with an estimated total usage of 1.33×10^6 tonnes (Voldner and Li 1993, Voldner and Li 1995). In 1984, the World Health Organization discouraged the use of this chemical on the basis of potential carcinogenic effects to humans, expressed reservations about the safety of this chemical in food, and considered it "a major hazard for aquatic and also some terrestrial species" (World Health Organization 1984). Despite this, toxaphene was used in some places for as much as another decade.

According to the strength of emissions and dominant wind circulation, toxaphene was transported by atmospheric pathways and deposited everywhere in North America (Rice and Evans 1984, Rapaport and Eisenreich 1986). Toxaphene concentration in air and water at Resolute Bay, Northwest Territories, was measured during August 1992 (Bidleman et al. 1995). Recent measurements of concentrations in sediment cores from Arctic lakes have shown that toxaphene was the major organochlorine pesticide accumulated in the lake sediments (Muir et al. 1995). Lake Superior's food web was analyzed in 1994 for hydrophobic organochlorine contaminants including toxaphene. Toxaphene was the dominant oganochlorine in the Lake Superior food web, with concentrations two to 15 times higher than total PCBs (polychlorinated biphenyls) (Kucklick et al. 1998). Toxaphene has also been measured in seawater from large areas of the Arctic Ocean (Jantunen and Bidleman 1998).

Contamination in the Arctic by persistent organochlorine pesticides is not a local issue, and the key to reducing the transport of these contaminants to the Arctic is not in the hands of Canadian policy makers. The direct connections between worldwide usage of HCH (hexachlorocyclohexane) and the air concentration of HCH in the Arctic has already been clearly identified (Li et al. 1998a). The atmospheric modelling community has been working to understand the occurrence and pathways of the contaminants. Goals are to differentiate between contributions from past use and from present sources, and to predict how these inputs and their concentrations in the various environmental compartments may change in the future. The model results will vary significantly depending on the available usage and emission data, and how these data are distributed globally.

The aim of this project is to compile information and create a computerized database of historical, present, and predicted global usage and emissions of toxaphene. Since the United States was the major user of toxaphene, and the total usage in this country reached > 40% of the total global usage, it is very important to create inventories of usage and emissions for toxaphene in the United States with more detailed resolution. In particular, use and emissions trend information is needed to interpret long-term trends of toxaphene in the Canadian Arctic environment.

Emissions can take place from contaminated soil and water. Agricultural soil is a large reservoir of toxaphene and likely a major source of emissions. Toxaphene levels in South Carolina and Alabama air during the mid-1990s were 10 times higher than near the Great Lakes and showed no relationship to air transport direction (Harner et al. 1999, Bidleman et al. 1998, Jantunen and Bidleman 1998). This suggests volatilization from regional soils that contain high toxaphene residues. Thus, it will also be very useful to create inventories for residues in agricultural soil in the United States. The goals of this research are to create gridded toxaphene emission inventories worldwide, including the major user, the United States, and to study the linkage between the toxaphene emissions and the toxaphene concentration in the Arctic air.

Activities in 1999-2000

Production, usage and geographic distribution

A large selection of literature and reports were searched and studied to collect information about the production and usage of toxaphene in the United States. Its usage on different crops and livestock has been identified, and usage in different farm production regions and different states has been allocated.

Usage gridding

In order to grid pesticide usage data, sets of surrogate data are needed to apportion the usage data. Gridded cropland along with cotton and soybean lands in the USA with 1/6° latitude by 1/4° longitude resolution have been created. These datasets have been compiled using an advanced very high resolution radiometer (AVHRR) global land database available from World Wide Web server for the Earth Resources Observation Systems (EROS) Data Center Node (United States Geological Survey 2000).

The gridded cattle number data set is used as a surrogate for apportion of toxaphene usage on livestock. A data set of cattle and calves number in the United States based on counties in 1987 from the 1992 Census of Agriculture (United States Census Bureau 2000) was used to create a gridded cattle number data set with the National Acid Precipitation Assessment Program (NAPAP) grid system. The census of agriculture is the leading source of statistics about the American agricultural production and the only source of consistent, comparable data at the county, state and national levels.

Emission inventories

According to our knowledge, emission inventories of toxaphene have never previously been reported. It is apparent that the computation of emissions on a grid system covering the whole United States requires considerable human and computer resources. The usage data, the annual emission factors and the application modes have been localized on a 1/6° latitude by 1/4° longitude grid system, and combined to estimate toxaphene emissions to the atmosphere. For highly persistent insecticides, such as toxaphene, it would be necessary to consider the emissions due to several consecutive years of application. For the present study, gridded usage of toxaphene from 1947 to 1985 has been used for estimating both emissions and residues for toxaphene. The resulting emission and residue estimates not only account for the present year's application but also any residual pesticide in the soil due to any application started in 1947.

Residues of toxaphene in agricultural soil

Methods for directly measuring the flux of pesticides from soil are well established (Majewski et al. 1993), but are labour intensive. Models are also available for predicting pesticide volatilization and emissions (Harner et al. 1998, Scholtz et al. 1997). In principal, such models could be used to predict emissions on a regional basis, but their application is limited by the paucity of pesticide residue data for soils. Our model calculates the residue of the toxaphene in the cropland.

Linkage between toxaphene emissions in the United States and the toxaphene concentration in the Arctic air

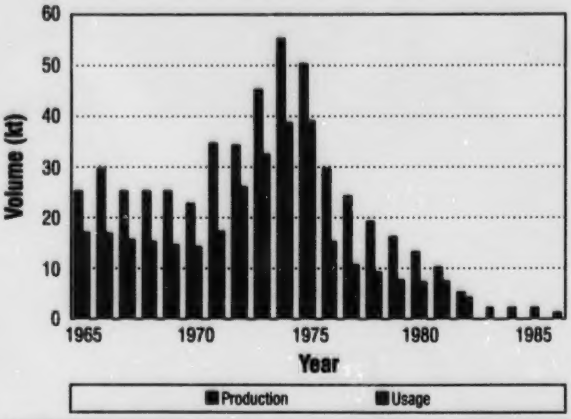
Linkage between toxaphene emissions in the United States and the toxaphene concentration in the Arctic air has been carefully studied. Atmospheric concentrations of toxaphene in the Arctic seem to be responding to reductions in toxaphene emissions from soil in the United States. This shows that toxaphene residues in American agricultural soil could be a major source of toxaphene in the Arctic air.

Results

Usage

The total toxaphene usage in the United States is about 490 kt, around 42% of the total global usage. The total production in the United States is 720 kt, around 230 kt were exported to other countries since 1946. Figure 1 presents production and usage of toxaphene in the United Sates between 1965 and 1986.

Figure 1. Annual production and usage of toxaphene in the United States, 1965-1985



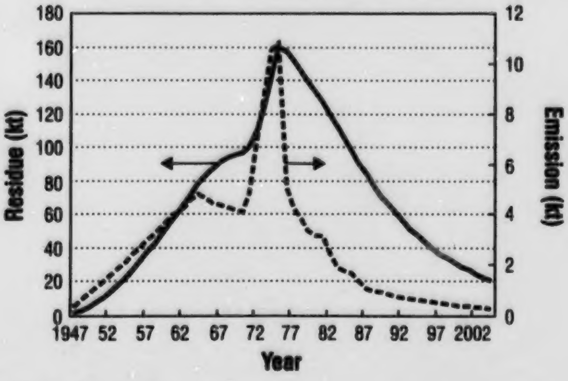
Trends of toxaphene emissions and residues in soil

Figure 2 shows the temporal trends of toxaphene emissions and residues in agricultural soil in the United States between 1947 and 2004. It indicates that both emissions and residues reached highest values in 1975 (160 kt for soil residues and 11 kt for emissions). Total toxaphene emissions are 194 kt between 1947 and 2000, 80% of which is contributed by the use of toxaphene in agriculture and 20% by its use on livestock.

Gridded toxaphene emissions and residues in soil

In 2000, almost 20 years after banning the use of toxaphene, there are still approximately 30 kt of toxaphene left in agricultural soil in the United States, and 360 t of these toxaphene residues in the agricultural soil will emit

Figure 2. Trends of toxaphene emissions and residues in agricultural soil in the United States, 1947-2004*



* Note the different scales.

to the air in the same year. The results are shown in Figure 3 for residues and Figure 4 for emissions.

Comparison with monitoring data: soil concentration

Data for pesticide application were collected in 37 states during the National Soils Monitoring Program, carried out in 1971 (Carey et al. 1978). Another survey was made of 36 Alabama agricultural soil samples in 1996 (Harner et al. 1999). The results from these two surveys are listed in Table 1. The table shows that the arithmetic mean of toxaphene concentration in the soil in 1971 is higher than that in 1996; however, the geometric mean in 1971 is lower than that in 1996. This indicates that toxaphene residues in 1996 were spread out from the source regions where toxaphene was used around 10 years ago due to transport and deposition.

Since the residue data from our calculation only present the concentration of toxaphene in the agricultural soil where toxaphene was applied, data from samples in Group 4 in the 1996 survey (Harner et al. 1999) are chosen for comparison. Also, we re-calculated the arithmetic mean of toxaphene concentration data for 1971 in Table 1 by averaging the total value among only those positive samples. These results are shown in Figure 5 along with the trends of usage, emissions and residues of toxaphene in agricultural soil in one grid cell close to Montgomery, Alabama, between 1950 and 2000. The curve of residues matches the monitoring data satisfactorily.

Figure 3. Toxaphene residues in the United States on a 1/6° latitude by 1/4° longitude grid system, 2000

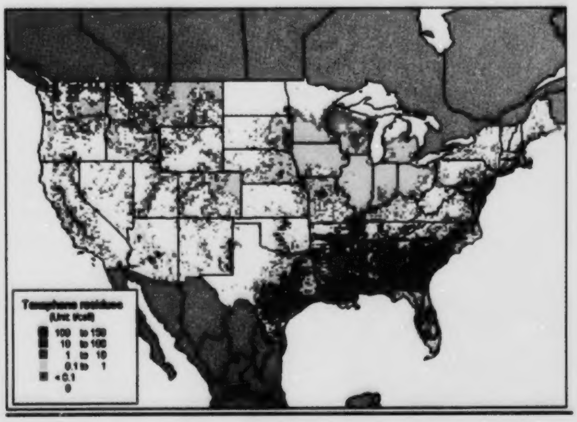
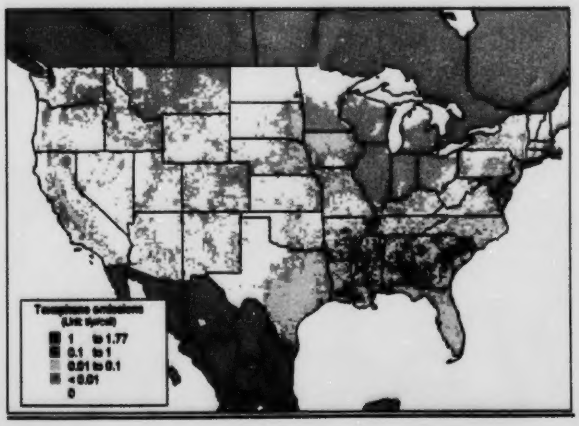


Figure 4. Toxaphene emissions in the United States on a 1/6° latitude by 1/4° longitude grid system, 2000



Comparison with input function derived from dated peat cores

Dated peat cores from Minnesota east to Nova Scotia have been analyzed for toxaphene (Rapaport and Eisenreich 1986). The composition of toxaphene in these cores indicated that the organic residues are delivered to the peat surface by atmospheric transport and deposited in a largely untransformed state; little transformation occurs once deposited in the peat. Using calculated accumulation rates in peat cores, the historical atmospheric input function for toxaphene from 1947 to 1985 is constructed and shown in Figure 6 along with total emissions of toxaphene from the United States. This input function is consistent with toxaphene emission data from the United States.

Table 1. Toxaphene residues (ng·g^{-1} dry weight) in Alabama soil samples

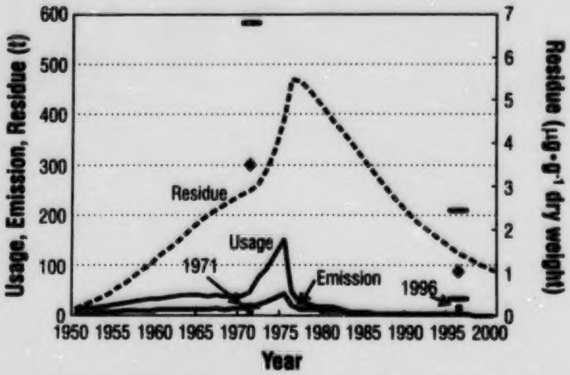
	1971*	1996**
No. of samples	23	39
Description of soil samples	Cropland	Private farm, experimental station and cemetery
No. of sites where toxaphene was used	3	NA***
% of sites where toxaphene was used	13.6	NA
No. of positives	5	38
% of positives	21.7	97.4
Minimum	180	< 3
Maximum	6780	2423
Arithmetic mean	760	285
Geometric mean	22	84
Detection limit	10	3

* Source: Carey et al.

** Source: Harner et al.

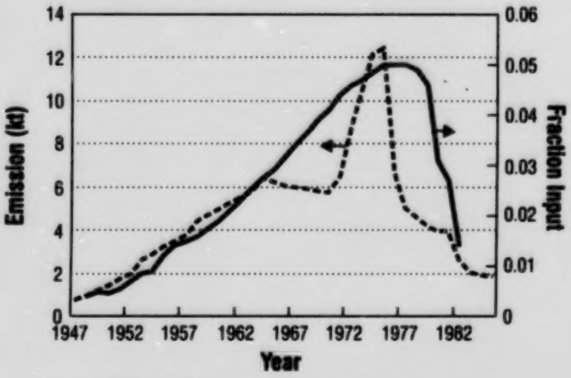
*** Not available

Figure 5. Trends of toxaphene usage, emissions and residues in agricultural soil in one grid cell in Alabama, 1950-2000*



* The measurements of soil concentrations of toxaphene (minimum, maximum and mean) in 1971 (Carey et al. 1978) and 1996 (Harner et al. 1999) are also presented. Note the different scales.

Figure 6. Total emissions of toxaphene from the United States and toxaphene atmospheric input function derived from peat accumulation in eastern North America, 1947-1985*



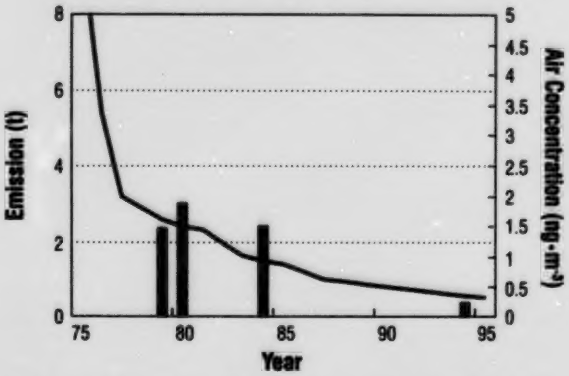
* Source of input function: Rapaport and Eisenreich 1986

Comparison with monitoring data: air concentration

Trends of toxaphene in ambient air of Columbia, South Carolina

A long-term record of toxaphene air concentration measurements was made in Columbia, South Carolina, and some results measured in October for different years are presented in Figure 7, along with the total annual emissions of toxaphene from the grid cell that contains the sampling site. It shows that the total toxaphene emissions match the air concentration data quite well. The air concentration in 1994 is quite low, since the higher emissions of toxaphene from the soil residues happened in plowing season (most likely May), not in October.

Figure 7. Toxaphene air concentrations (bars) in October in Columbia, South Carolina, and the total emissions of toxaphene (curve) from the grid cell that contains the sampling site*

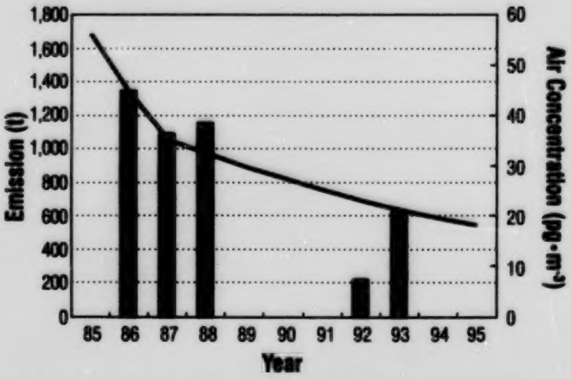


* Sources for air concentration: 1979 and 1980: Billings and Bidleman 1983; 1984: Foreman and Bidleman 1987; 1994: Bidleman et al. 1998

Trends of toxaphene in Arctic air

Although the number of samples from the 1980s was small, recent atmospheric measurements suggest that a decline in toxaphene has taken place (Arctic Monitoring and Assessment Program 1998). Figure 8 shows the temporal trends in concentrations of toxaphene in the Arctic air (the bars) and the emissions of toxaphene from the United States (the curve) from 1985 to 1995. It indicates that the toxaphene air concentrations in the Arctic are well correlated with emissions from the United States.

Figure 8. Temporal trends in concentrations of toxaphene in the Arctic air (bars) and the emissions of toxaphene from the United States (curve), 1985-1995*



* Source of measurement: Arctic Monitoring and Assessment Programme 1998

Discussion and Conclusions

Atmospheric concentrations of toxaphene in the Arctic seem to be responding to reductions of toxaphene emissions from soil in the United States. This indicates that toxaphene residues in the United States could be a major source of toxaphene in the Arctic air. A continuation in the trend in declining air concentrations in the Arctic is expected as this compound decreases in soil in the United States due to emission and degradation. However, since toxaphene concentrations in Arctic waters have remained relatively unchanged, the net direction of air-sea gas flux could be reversed as the air toxaphene concentration drops. Thus, the Arctic waters containing toxaphene will become a source to the atmosphere that will likely maintain detectable atmospheric concentrations for some time in the future.

Project Completion Date

May 31, 2000

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Mercury and Other Contaminants in a Laminated Sediment Core from Devon Island

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Abstract

There has been debate in the scientific literature regarding whether lake sediment records provide an accurate surrogate for histories of the deposition of mercury. One source of ambiguity in sediment records is physical mixing of sediments due to the processes like wave action, ice scouring and activities of benthic animals. The reports of laminated sediments in lake DV09 on Devon Island, Nunavut, offered the possibility to examine the apparent history of deposition of mercury in the absence of artifacts of physical mixing of sediment layers. Four cores were taken from the lake in May 1999. Core 1 has been analyzed for dating radioisotopes, carbon forms, several metals, including stable lead isotopes, and some organic contaminants. The results indicate a history of low but consistent increases in inputs of mercury but offer no evidence of increased inputs of industrial lead. Consistent with an atmospheric source of contaminants to this remote lake is the detection of ^{137}Cs from bomb testing and from Chernobyl and also a variety of synthetic organochlorine compounds.

Objectives

1. To determine the down-core profiles of mercury and several other contaminants in annually laminated sediments from Lake DV09 on Devon Island, Nunavut.
2. To test the argument that inputs of several elements due to long-range transboundary air pollution can be detected in a remote lake where sediment mixing does not confound the core interpretation.
3. To calculate modern and historic fluxes of contaminants to the site.

Introduction

Several independent lines of evidence suggest that recent inputs of mercury (Hg) and a number of other materials to northern Canada are greater than those of pre-industrial times. These arguments are unambiguous in instances of synthetic elements and compounds with no natural sources. For example, cesium-137 (^{137}Cs) has no natural source but occurs exclusively as a by-product of nuclear fission, a process not known before the 20th century. It was released into the air by atmospheric testing of nuclear bombs and it is still detected consistently throughout the Arctic (e.g. Lockhart et al. 1998). Synthetic organic compounds like chlordane and toxaphene are products exclusively of the last half-century and they, too, are consistently found in the Arctic although they were not used there. The distribution of these synthetic materials leaves no doubt that contaminants make their way to even the most remote areas of the Arctic.

With natural elements, the problem of calculating fluxes and of discriminating natural from anthropogenic influences is more difficult since they occur naturally and, in addition, some of them have been dispersed throughout the environment as a result of human activities. The strongest evidence for this is the element lead (Pb) used for several decades as an additive to gasoline. Hg is the most problematic of the metals in Arctic ecosystems because it accumulates in aquatic food chains and reaches levels in some animals which exceed Health Canada and World Health Organization levels of concern for consumption.

Lake sediment cores record inputs of materials to lakes. Core records have been used to compare modern and historic inputs of Hg and Pb in a number of settings, including the Arctic. Sediment cores are subject to several kinds of artifacts that can make interpretation difficult. One such artifact is physical mixing of sediments by processes such as wave action, ice scouring and movements of benthic animals. In some instances sediments are deposited in visible layers. When that occurs, physical mixing must be essentially non-existent or the layer structure would not have been preserved. Lake DV09 on northern Devon Island is such a lake (Gajewski et al. 1997). This project was conducted to test whether an Arctic lake with evidence of little or no physical mixing would contain a core profile showing a gradient of Hg with more in the upper, recent layers than in the lower, older layers.

Activities in 1999-2000

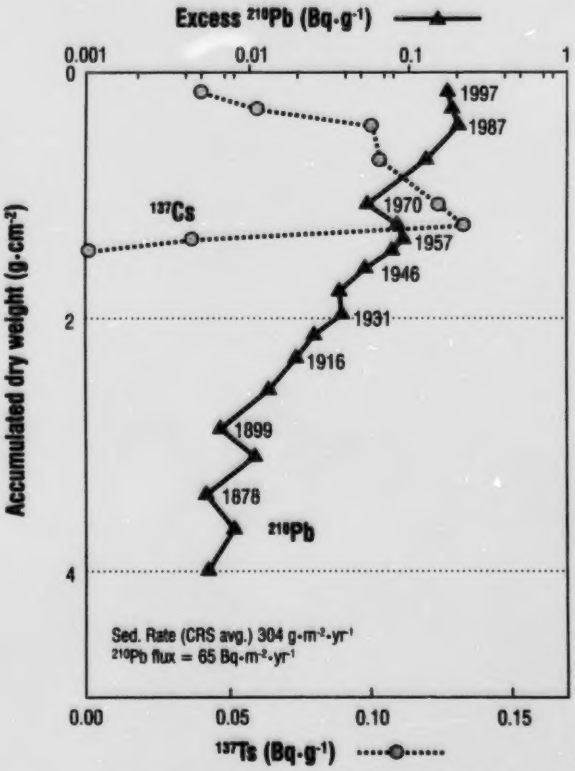
An expedition to Lake DV09 was made in May 1999, using a ski-equipped Twin Otter from the Polar Continental Shelf Project in Resolute. Four cores were collected, approximately 4 m apart at 75° 34.420´N latitude, 89° 18.545´W longitude. The core tops were all undisturbed and distinct laminations were observed down the lengths of each. Cores 1 and 2 were sliced in 0.5 cm and 1 cm intervals at the core site and cores 3 and 4 were sealed intact in the core tube with plastic and styrofoam plugs. The two sliced cores were brought to the Freshwater Institute in Winnipeg, Manitoba, for dating and chemical analyses, and the two intact cores were brought to the Geological Survey laboratories in Ottawa, Ontario, for geological and geochemical analyses. Here we report results to date for core 1.

Results and Discussion

The cores were all visibly laminated, confirming that physical mixing of sediment particles was essentially non-existent in these sediments.

The down-core profiles for dating radioisotopes in core 1 are shown in Figure 1. The profile for lead-210 (^{210}Pb) shows an exponential decline in the amount of this isotope over time and the median dates of deposition for representative slices are indicated on the graph. One of the tests of the credibility of the dating is whether or not the ^{210}Pb dates for the early 1960s coincide with the peak inputs of ^{137}Cs. ^{137}Cs is a fission product which was released by atmospheric testing of nuclear bombs. The peak year for bomb testing was 1963 and that year is indicated on the graph as the year when maximum inputs of ^{137}Cs occurred (red). There was a second small shoulder on the ^{137}Cs peak in the slice dated 1987. That

Figure 1. ^{210}Pb and ^{137}Cs activities in slices of core 1 from Devon Island Lake DV09, May 1999*



* Estimated dates of deposition of mid-points of slices are shown on the ^{210}Pb profile.

probably represents a small input of ^{137}Cs from the Chernobyl reactor accident in Ukraine in 1986. Input from Chernobyl was also reported by Lockhart et al. (1993) in sediments from Hawk Lake (Saqvaqjuac). In the study of a core from the same lake, Gajewski et al. (1997) found that the ^{210}Pb dates gave ages consistent with counts of laminations. We have not yet made the comparisons between laminations and Pb dates in the cores taken in 1999, but we hypothesize results consistent with the earlier core. Taken together, the exponential decline in the ^{210}Pb profile and the timing of the ^{137}Cs inputs indicate a core with a credible historical record. The sedimentation rate calculated from the ^{210}Pb data was 304 $g \cdot m^{-2} \cdot y^{-1}$. There was some focusing of sediment to the site because the flux of ^{210}Pb was 65 $Bq \cdot m^{-2} \cdot y^{-1}$, a value higher than expected for 75° N latitude. We have used soil profiles to obtain ^{210}Pb fluxes of 60 $Bq \cdot m^{-2} \cdot y^{-1}$ at Saqvaqjuac (63°N) and only 7 $Bq \cdot m^{-2} \cdot y^{-1}$ at Lake Hazen (82°N) (Lockhart et al. 1998) and the DV09 site should fall between them. Recently we obtained a flux of 27 $Bq \cdot m^{-2} \cdot y^{-1}$ for a soil profile taken from the north shore of Banks Island in 1998 (74°N). The flux at Lake DV09 should be similar to that for the Banks Island site since they were at essentially the same latitude. Assuming an expected flux of ^{210}Pb of 27 $Bq \cdot m^{-2} \cdot y^{-1}$, the actual flux of 65 $Bq \cdot m^{-2} \cdot y^{-1}$ indicates that the measured flux is too high by a factor of 2.4 (65/27). This correction is due to focussing of sediment to the sampling point such that it has received more ^{210}Pb than that expected for its surface area.

Several metals measured in the core slices are shown in Figure 2. The metal of particular interest for this project is Hg. Hg concentrations increased consistently in the upper slices dating from about the 1920s. The original question responsible for this study was whether a gradient in Hg concentration would be found in laminated sediments. The Hg profile shown in Figure 2 leaves no doubt that a gradient in Hg can be produced in the absence of particle mixing. We reported similar profiles from a number of northern lakes (Lockhart et al. 1993, Lockhart et al. 1995, Lockhart et al. 1998), as did Ouellet and Jones (1982) and Lucotte et al. (1995) from northern Quebec, and Hermanson (1993) from the Belcher Islands.

The flux of Hg can be calculated from the product of the concentration and the sedimentation rate, and this product can then be adjusted for focussing. The concentrations can be taken for each slice or from any arbitrary group of them. Using the same procedure of Lockhart et al. (1998) the average concentration for the slices deposited since 1950 (slices 1 to 8) was calculated as 0.027 $\mu g \cdot g^{-1}$. Combining that with the sedimentation rate of 304 $g \cdot m^{-2} \cdot y^{-1}$, an uncorrected Hg flux of 8.2 $\mu g \cdot m^{-2} \cdot y^{-1}$ is obtained. Then, dividing by the focus factor of 2.4, a corrected flux of Hg of 3.4 $\mu g \cdot m^{-2} \cdot y^{-1}$ is calculated as the average for the period between 1950 and 1999. Repeating the same calculation for the deepest five slices, the average Hg concentration was 0.015 $\mu g \cdot g^{-1}$ and the corrected flux was 1.7 $\mu g \cdot m^{-2} \cdot y^{-1}$. Hence the recent flux was 3.4 $\mu g \cdot m^{-2} \cdot y^{-1}$ and the historic flux was 1.7 $\mu g \cdot m^{-2} \cdot y^{-1}$. The difference between these two figures is 1.7 $\mu g \cdot m^{-2} \cdot y^{-1}$ and we attribute that to anthropogenic inputs. We lack a credible alternative explanation for the increased fluxes. We have relatively good information on anthropogenic emissions of Hg and deposition to the surface of the earth is needed to balance the inputs and the atmospheric concentrations. Furthermore, there are data now showing concentrations of Hg in Arctic air (Schroeder et al. 1998) and there is also evidence that Arctic snow contains relatively high levels of Hg (Lu et al. 2000, manuscript submitted for review). Consequently, a mechanisms to deliver Hg from emission sources to the Arctic is established.

Of the remaining metals shown in Figure 2, Pb has received the most study in sediments. Sediments at mid-latitudes show distinct peaks of Pb in slices dated from the early 1900s due largely to the addition of alkylated Pb to gasoline (see Norton et al, 1989 for a review). Hermanson (1993) observed a clear increase in Pb in lake sediments from the Belcher Islands. We have seen an increase in Pb in sediments from Far Lake on the west coast of Hudson Bay at 64°N, at Sophia Lake on Cornwallis Island (75°N) and possibly at Amituk Lake (also 75°N), near Sophia Lake (Figure 3). The extreme northern lakes (Hazen and Buchanan) had erratic Pb profiles with no clear trend. There may be a slight downward trend in Lake Hazen sediments and a slight upward trend in those from Buchanan (Figure 3). Further south, sediments of Lake 375 in northwestern Ontario have recorded a dramatic increase in Pb.

The Pb profile from lake DV09 showed considerable variation from slice to slice with a subtle increase from deeper to surficial layers (Figure 2). Analyses for stable Pb isotopes revealed no evidence of anthropogenic Pb, as evidenced by the lack of change in ^{206}Pb/^{207}Pb isotope ratios between the bottom and top of the core (Figure 4). We examined a core from Clay Lake in northwestern Ontario for Pb isotopes and found definitive evidence of isotope ratio changes consistent with inputs from leaded gasoline (Lockhart et al. 2000, in press). The Arctic atmosphere contains a ^{206}Pb/^{207}Pb ratio of about 1.16, which is largely European and Eurasian industrial Pb. If even 20% of the core Pb was from this source, we should have seen a shift of about 1.5% in the ^{206}Pb/^{207}Pb ratio. The North American atmospheric industrial signatures are either 1.16 (Canada) or 1.20 (United States). There was a shift in the ^{208}Pb/^{207}Pb data of about 1.2%, but it is not possible in this context to introduce industrial Pb and

Figure 2. Down-core profiles of several metals and forms of carbon in slices of sediment from core 1 from Lake DV09, Devon Island, May 1999

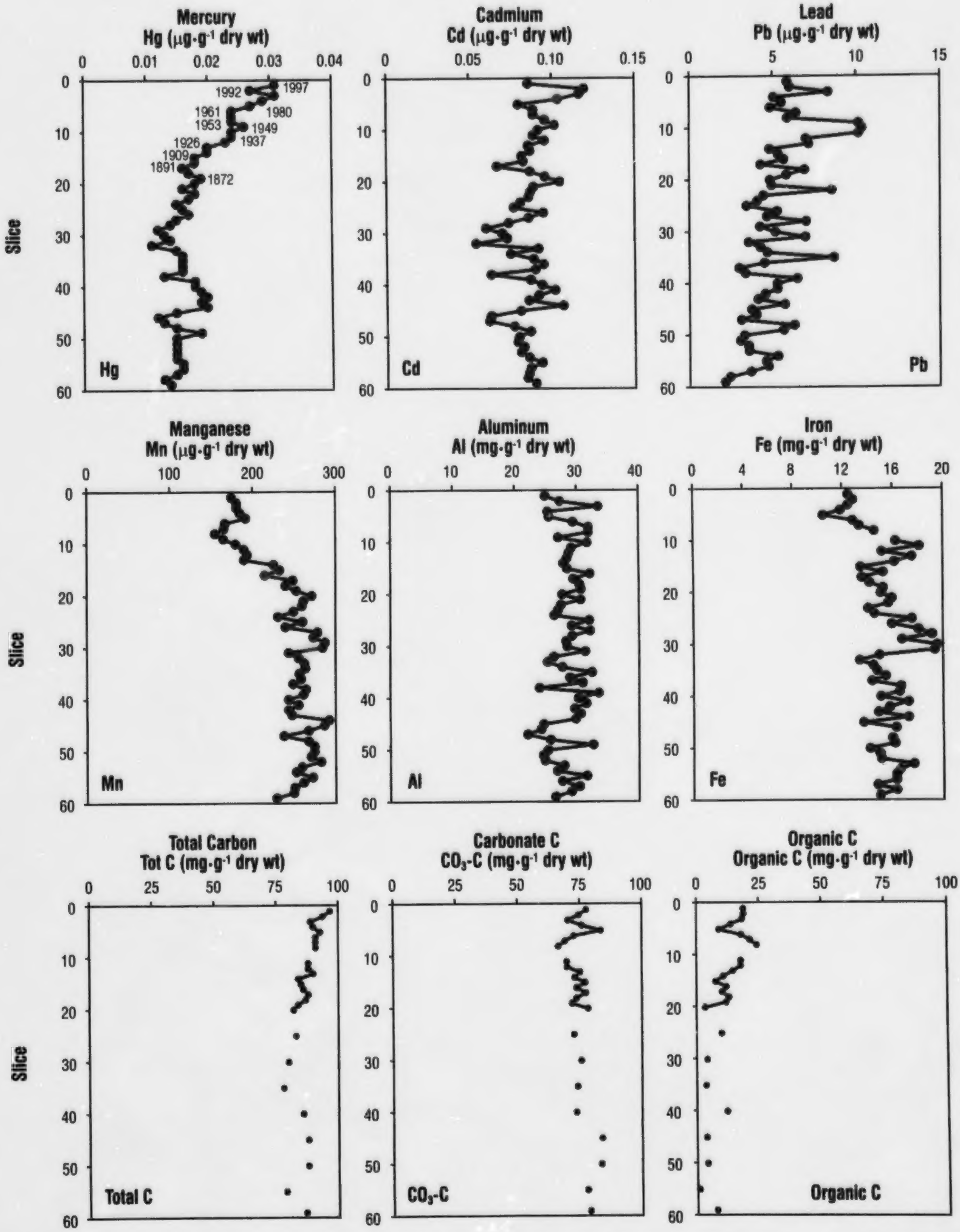
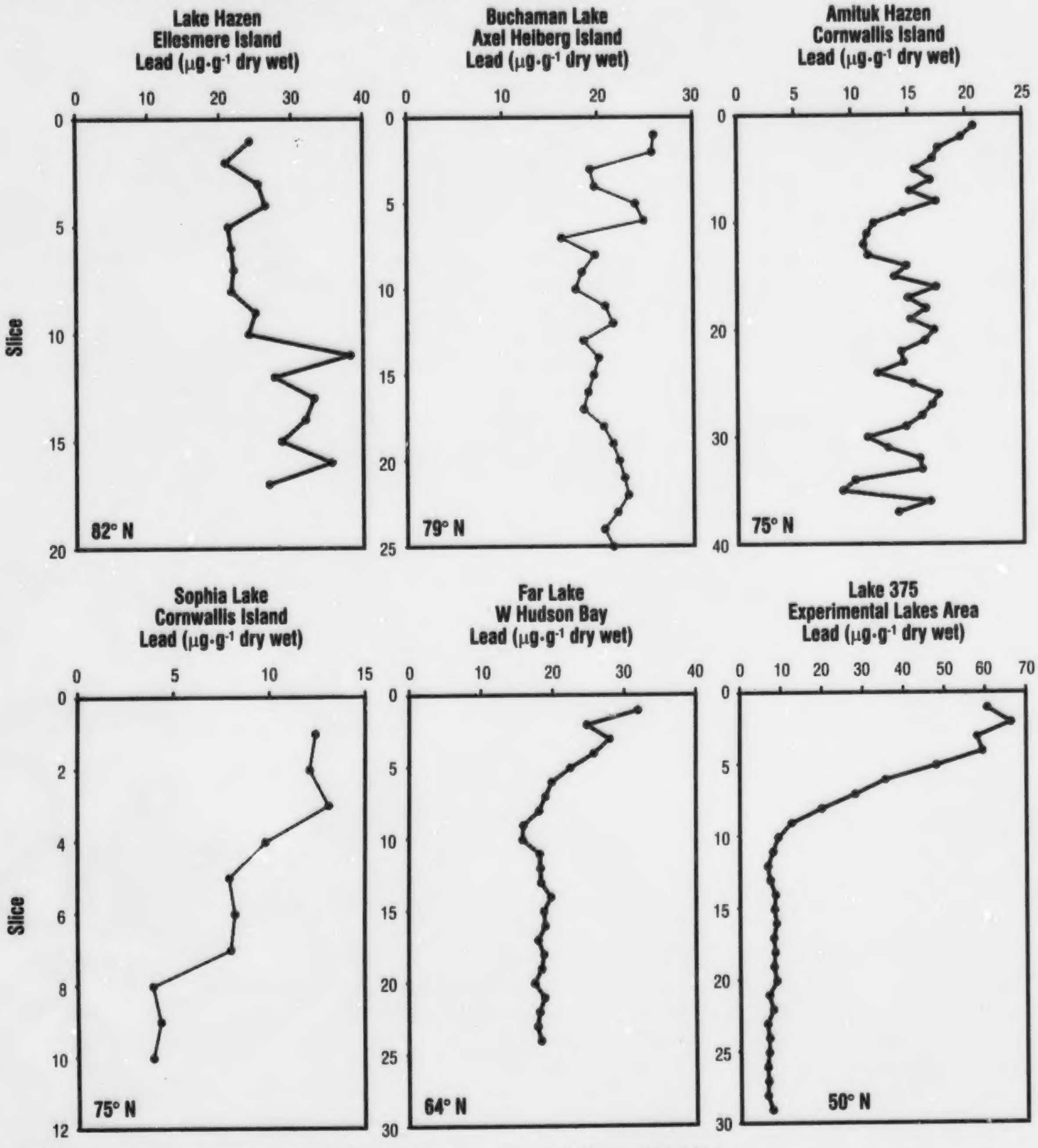


Figure 3. Down-core profiles of lead in sediment cores from several lakes*



Lake Hazen
Ellesmere Island
Lead (µg·g⁻¹ dry wet)

Buchaman Lake
Axel Heiberg Island
Lead (µg·g⁻¹ dry wet)

Amituk Hazen
Cornwallis Island
Lead (µg·g⁻¹ dry wet)

Sophia Lake
Cornwallis Island
Lead (µg·g⁻¹ dry wet)

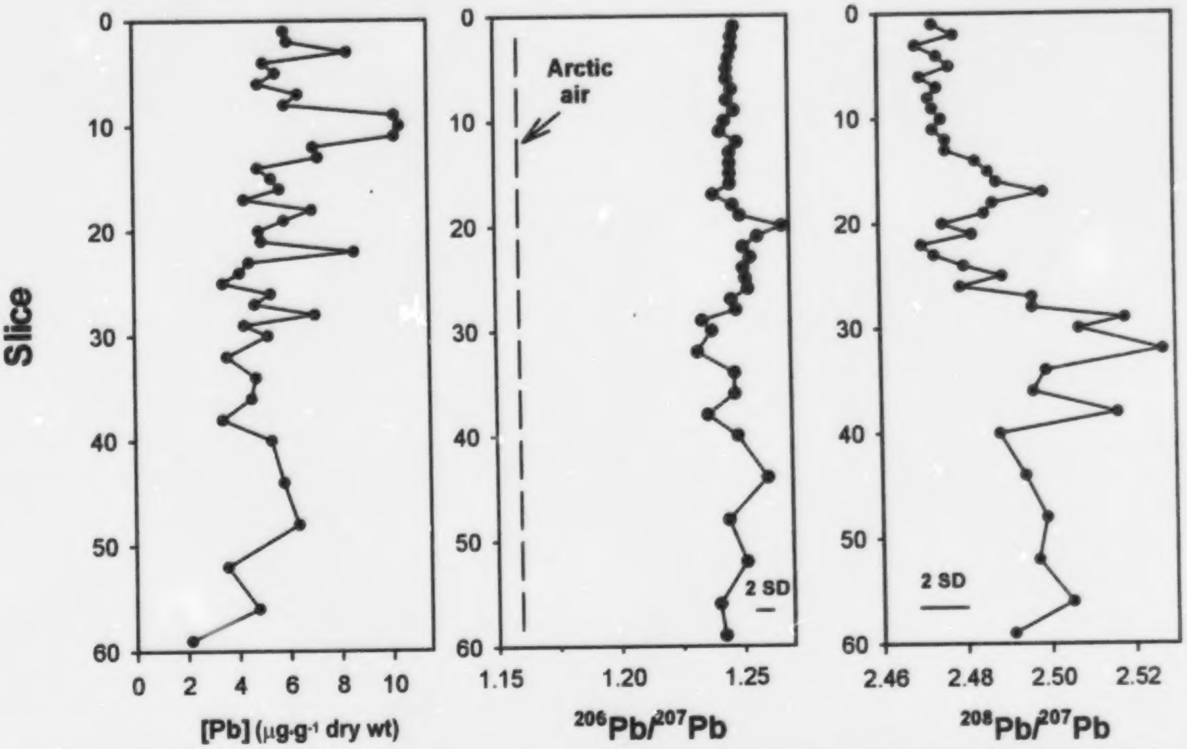
Far Lake
W Hudson Bay
Lead (µg·g⁻¹ dry wet)

Lake 375
Experimental Lakes Area
Lead (µg·g⁻¹ dry wet)

* The approximate latitude of each lake is shown in the lower left corner of each panel (Source: L. Lockhart, unpublished data).

get a change in one ratio but not the other. The change in $^{208}Pb/^{207}Pb$ and the weak trend towards increased Pb concentration in the upper slices may reflect changing local conditions; perhaps reduced snow cover leading to more weathering in the drainage in recent times. With weathering, fractionation of Pb isotopes has been observed, as different mineral phases weather at different rates. That may be what is happening on north Devon Island to produce a slight shift in $^{208}Pb/^{207}Pb$ but not in $^{206}Pb/^{207}Pb$. Gajewski et al. (1997) interpreted the varve structure and diatom remains as indications that Lake DV09 has experienced longer ice-free periods or warmer summer temperatures in the last half-century than in earlier times. However, the profile for total Pb (Figures 2 and 4) did not show an increase in the last half-century but rather

Figure 4. Down-core concentrations of lead ($\mu g \cdot g^{-1}$ dry wt) (left panel) in core 1 from lake DV09, ratio of $^{206}Pb/^{207}Pb$ (centre) and $^{208}Pb/^{207}Pb$ (right panel)



an erratic profile with an apparent long-term but very small increase over the whole period sampled by the core.

The weathering hypothesis suggests that not only Pb but other elements also could be reaching the lake in increased quantities due to weathering. This suggests a likelihood of correlations among them and indeed several such correlations were observed (Table 1). Down-core concentrations of Hg was correlated with those of cadmium (Cd) ($r = 0.66$) and Pb ($r = 0.55$) but more strongly with carbon, notably with total carbon ($r = 0.73$) and organic carbon ($r = 0.73$) as opposed to the carbonate ($r = -0.31$, not significant). Hg was correlated most strongly with nitrogen ($r = 0.78$, not shown in Figure 2). The total carbon increased only slightly (range: 78 to 97 $mg \cdot g^{-1}$) over the depth of the core, and so the correlation with total carbon reflects the relationship between Hg and organic carbon (range: 0.8 to 19 $mg \cdot g^{-1}$). The gradient in organic carbon is unlikely to be the cause of the gradient of Hg in the sense of driving a redistribution of Hg within the core. Lucotte et al. (1995) examined a number of cores from central Quebec and found that most had little or no gradient between carbon at the top and carbon at the bottom but that Hg gradients were found anyway. It may be that recent warming has resulted in increased inputs of organic carbon from the watershed and also has enabled more production of organic carbon within the lake. The strong relationship between Hg and nitrogen may represent some mixture of the same phenomena. We might expect an increase in weathering to result in increased supplies of organic matter to the lake and with that, any Hg associated with it. Given the high affinity of Hg for organic matter, increases in the supply of Hg from the air would be expected to result in inputs of Hg bound to organic matter in the watershed or in the lake and ultimately to increased supplies of organically bound Hg to the lake.

Iron (Fe) and manganese (Mn) (Figure 2) are often difficult to interpret in sediment cores because of their complex relationships to oxygen. These elements form soluble species under anaerobic conditions but form insoluble species in aerobic conditions. The only profile for oxygen available for Lake DV09 is that of Gajewski et al. (1997) who obtained a profile from a site 14 m deep on July 21, 1994, when the lake was partially covered with ice; oxygen was in the range of 12.7 to 13 $mg \cdot L^{-1}$ throughout. Fe and Mn both show declines in slices near the surface of the core but these changes seem unlikely to reflect declining inputs. One mechanism might be late

Table 1. Correlation coefficients for pairs of metals and carbon forms, core DV09-1, May 1999

		Hg	Cd	Pb	Mn	Al	Fe	Ti	Total C	CO_3	Organic C
Hg		1.000									
	p	0.000									
	n	59									
Cd		0.657	1.000								
	p	0.000	0.000								
	n	59	59								
Pb		0.546	0.366	1.000							
	p	0.000	0.004	0.000							
	n	59	59	59							
Mn		-0.809	-0.397	-0.474	1.000						
	p	0.000	0.002	0.000	0.000						
	n	59	59	59	59						
Al		0.183	0.363	0.368	-0.108	1.000					
	p	0.165	0.005	0.004	0.416	0.000					
	n	59	59	59	59	59					
Fe		-0.295	-0.096	0.283	0.259	0.246	1.000				
	p	0.023	0.468	0.030	0.047	0.061	0.000				
	n	59	59	59	59	59	59				
Ti		-0.086	0.156	0.122	0.068	0.524	0.152	1.000			
	p	0.519	0.238	0.359	0.611	0.000	0.250	0.000			
	n	59	59	59	59	59	59	59			
Total C		0.728	0.200	0.067	-0.756	-0.395	-0.603	-0.519	1.000		
	p	0.000	0.327	0.744	0.000	0.046	0.001	0.007	0.000		
	n	26	26	26	26	26	26	26	26		
CO_3		-0.307	-0.333	-0.556	0.511	-0.707	-0.271	-0.392	-0.001	1.000	
	p	0.127	0.096	0.003	0.008	0.000	0.181	0.048	0.994	0.000	
	n	26	26	26	26	26	26	26	26	26	
Organic C		0.734	0.376	0.438	-0.897	0.215	-0.239	-0.095	0.713	-0.703	1.000
	p	0.000	0.058	0.025	0.000	0.292	0.240	0.646	0.000	0.000	0.000
	n	26	26	26	26	26	26	26	26	26	26

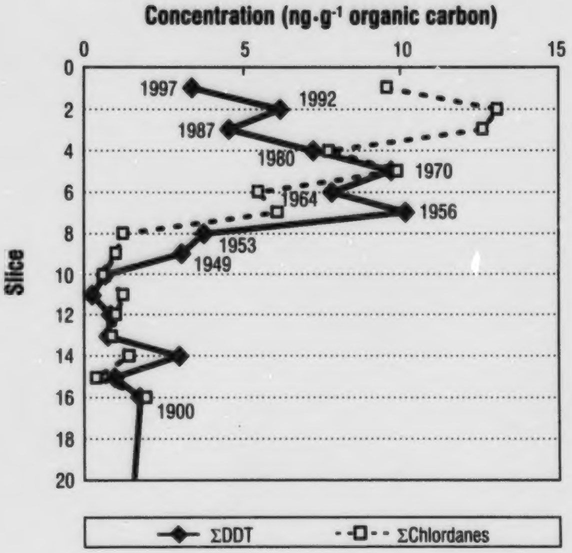
winter anoxia allowing the dissolution of Fe and Mn from the sediments back into the water column and export through the outlet.

Aluminum (Al) (Figure 2) and titanium (Ti) (not shown) usually track erosional inputs from the drainage basin and they would not be expected to change unless some major change had occurred in the drainage basin or the sedimentation rate.

Cd showed no clear gradient in the core although there may be some hint of small increases near the top. Ignoring the issue of redistribution of Cd within sediments, this core suggests that inputs of Cd have changed very little or not at all.

Some of the slices were also analyzed for a wide array of organochlorine compounds, a representative selection of which (ΣDDT and Σchlordane) is shown in Figure 5. These profiles, like the ^{137}Cs shown in Figure 1, show definitively that materials originating exclusively with human activities have reached the site. Furthermore, in view of the location of the site remote from any local human activity, the source of these synthetic materials must have been from atmospheric deposition. The shapes of the profiles are considerably sharper than those produced previously in the Northern Contaminants Program-sponsored project (e.g. Muir et al. 1996), which is likely the result of thinner slices taken from core DV09

Figure 5. Down-core profiles of of ΣDDT and Σchlordanes in slices of core DV09-1, May 1999*



* Concentrations have been normalized to the organic carbon content of each slice. Estimated median dates of slice deposition are shown on the ΣDDT (solid) line.

and lack of physical mixing in the laminated sediments. The increases were typically evident by about slice 9, which was dated to 1949 using the ^{210}Pb profile (linear fit).

Conclusions

The reason for choosing to sample lake DV09 was the presence of visibly laminated sediments, which precludes significant physical mixing. The question to be answered was whether a gradient in Hg concentration could have been produced in the absence of physical mixing. The core provided a clear answer: a gradient of Hg was established in spite of the absence of physical mixing.

Two major questions challenge the interpretation of Hg profiles in cores as histories of deposition. One is whether gradients in Hg might be by-products of physical mixing of sediment particles. If so, then the absence of physical mixing might imply the absence of a gradient in Hg. This core has effectively established that physical mixing could not have been the cause of the gradient in Hg.

A second question is whether gradients in Hg might be artifacts produced in response to other chemical gradients. This question cannot be answered definitively from this study and it was not intended to be. We have compared known Hg pollution histories from a number of sites in Canada against histories inferred from sediment cores and found that the cores recorded quite faithfully the known histories.

The lack of any evidence of anthropogenic inputs of Pb is of considerable interest. This will prompt the examination of some other lakes where there was clear evidence of increased inputs of Pb to determine whether the isotopic techniques show the increases to have been driven by sources outside the drainages.

The presence of ^{137}Cs from bomb testing and from Chernobyl, and also the presence of a variety of synthetic organochlorine compounds, shows that inputs of atmospherically transported materials have reached even this remote lake.

Expected Project Completion Date

Field activity was completed in May 1999. Analyses are continuing and should be completed for core 1 by December 2000. Work on core 2 will be undertaken only if questions arise from core 1 that require reference to the second sliced core. Geological and geochemical work on the unsliced cores is underway at the Geological Survey of Canada. Depending on the results of the geological examinations, these cores may offer several additional insights for further analytical work in the future.

Acknowledgments

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Review and Models of Contaminant Fate Designed to Support International Negotiations

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Abstract

This study involved three tasks. First has been the development of a more user-friendly version of the Wania-Mackay mass balance model of organic chemical fate on a global scale suitable for use by a variety of users, especially those from abroad. This model is now available for free downloading from a Web site. Second has been the development of an evaluative model for assessing the persistence and long-range transport potential of chemicals with particular emphasis on how these attributes are affected by cold climate conditions. Third has been the preparation of a "popular" article written at a level suitable for the informed lay public which describes the processes of chemical transport and accumulation in Arctic ecosystems. In general, the three tasks have the objective of contributing to improved understanding of how chemicals migrate to the Canadian Arctic and behave in Arctic ecosystems. The project provides documentation and support for Canadian international negotiations to reduce and eliminate the use of persistent organic pollutants which are of concern in the Canadian North.

Objectives

1. To develop a simplified version of the Wania-Mackay global model to be used in other countries to determine the extent to which specific chemicals emitted at various locations are likely transmitted into the Arctic. The model is to be fully documented and made available freely to all interested parties on a Web site.
2. To develop an evaluative model which will assess chemicals for their persistence and potential for long-range transport into the Arctic.
3. To prepare one or more "popular" articles at the *Scientific American*, *Discover* or *Canadian Geographic* level, with attractive and easily understood graphics, aimed at the lay public to explain the nature and importance of northern contamination.

Introduction

Canadian Government negotiations have played a significant role in international fora concerned with the global control of persistent organic pollutants (POPs). In

1999, the United Nations Environment Programme proposed a list of 12 substances or groups of substances for elimination or severe regulation, largely as a result of pressure from Canada, driven by concerns about "northern contaminants". It is likely that other substances will be similarly regulated in the future. These agreements often involve cost, inconvenience and adverse effects to nations that use these substances, thus agreement may be difficult to achieve. Notable examples are the use of DDT for malaria control in tropical countries, and the still widespread use of PCBs (polychlorinated biphenyls) in components of electrical power distribution systems.

International support for controls on POPs can be enhanced by providing a sound scientific basis for the proposed measures and demonstrating the quantitative linkage between use in the South and the consequent exposure in the North. In this study, a version of the Wania-Mackay global model of chemical fate has been modified to facilitate its use by scientists in other nations. This model has been peer reviewed, published (Wania et al. 1999a, Wania and MacKay 1999a) and well received but to date it has not been in a form in which scientists in other countries could apply. The first task has been to simplify the model implementation and provide a mechanism through the Internet for its international dissemination.

As part of these international POPs activities, criteria have been developed for persistence, bioaccumulation, toxicity and potential for long-range transport which can be used to assign priorities to lists of candidate POPs (Mackay et al. 2000). These assessments are normally done at 25°C, the temperature at which data are usually gathered. In this study we explore the issue of how low temperatures, as those that prevail throughout the Canadian North, may influence the priority rankings.

Support for international controls depends, in part, on public pressure which is derived from information provided through the news media, magazines and books. There is a need for a relatively simply stated justification for international controls of chemicals from the perspective of protecting the ecosystem and human health of the Canadian North. Part of this project has been the compilation of such a "popular" account of the issues in the hope that it may favourably influence public opinion and hence governmental attitudes and responses.

Activities in 1999-2000

Global model

The existing model was suitable only for use by the developer, largely because there was no detailed documentation of the parameter input procedures. Further, the modelling procedures themselves, while reliable, were not easily understood or implemented. Similarly, the outputs were not fully explained. The major activity has been the revision of the model to a form which can be relatively easily used by other individuals to make the results more readily interpretable and to document this in a user manual.

Persistence and long-range transport

Sound and robust methods of estimating persistence and potential for long-range transport have been developed and published with support from the Northern Contaminants Program (NCP) and other sources. Subsequently the effect of low temperature on these estimation methods has been explored for a suite of some 230 chemicals of environmental interest from our CD-ROM database (Mackay et al. 1999).

Popular article

This article has been fully researched using the refereed literature, various international contacts and advice from several workers who have contributed to the NCP program. A first draft has been written and graphics are being prepared. It is hoped that the article will be sent to potential publishers during the summer of 2000.

Results

The global model was successfully revised and has been made available through the Web site created at the University of Toronto by Dr. Frank Wania (www.scar.utoronto.ca/~wania/downloads2.htms). There is also a link from the Trent University Web site (www.trentu.ca/envmodel). Both the model and the manual (Wania and Mackay 2000) can be downloaded by accessing these sites.

Beta testing has been undertaken and has revealed a number of problems which are being resolved. In particular, the model is implemented in a Windows platform but not all versions of Windows can support it. The reasons for this are not fully understood. We are testing the model internationally at the Canadian Environmental Modelling Centre and are awaiting responses from other users.

Persistence and long-range transport criteria for POPs: role of low temperatures

With partial support from the NCP, two papers were written and published (Gouin et al. 2000, Beyer et al. 2000). In the first, we examined the existing methods of evaluating persistence, then developed a novel and simple approach. The effects of a 25°C drop in

Figure 1a. **Plot of Log K_{AW} versus Log K_{OW} at 25°C showing the distribution of 233 chemicals and lines of equal fractions in air, water and octanol***

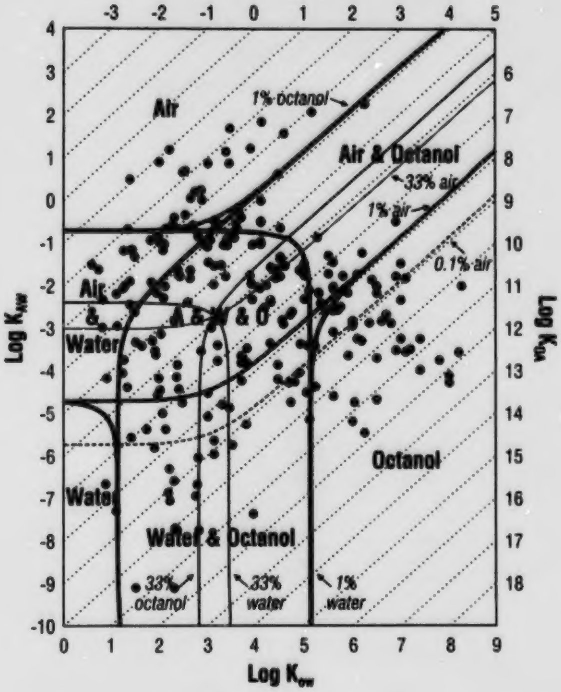
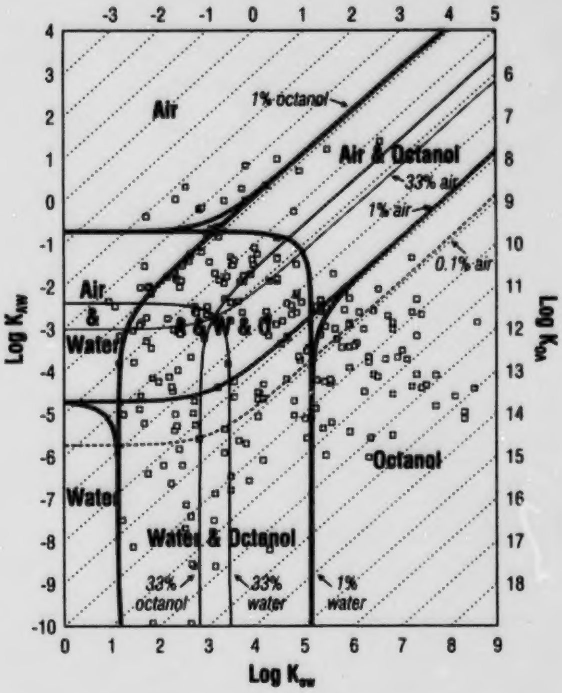


Figure 1b. **Plot of Log K_{AW} versus Log K_{OW} at 0°C showing the distribution of 233 chemicals and lines of equal fractions in air, water and octanol***



* Data from Mackay et al. 1999. The 45° diagonal lines represent constant log K_{OA} the octanol-air partition coefficient. The overall persistence of those substances in the upper left region, where more than 99% is in air, is controlled by the air half-life. Similarly, the overall persistence of those substances in the region to the lower right, where more then 99% is in octanol, is controlled by the octanol half-life, while those substances located in the region to the lower left, where > 99% is in water, has an overall persistence that is controlled by the water half-life.

temperature were then explored. In the second, we proposed a rational method of prioritizing chemicals according to their long-range transport potential using information from multi-media fate models as input. Part of this work was a detailed evaluation of the effect of temperature on long-range transport. This evaluation was not included in the paper for reasons of space limitation but the key findings which are given below will be the subject of a subsequent report and paper.

Persistence

It has been shown that persistence can be evaluated using a simple Level II fugacity model in which the environment is treated as defined relative volumes of air, water and octanol, the latter being a surrogate for organic matter (Gouin et al. 2000). From data on the substance's air-water (K_{AW}) and octanol-water (K_{OW}) partition coefficients at 25°C, the proportions in each phase at equilibrium can be deduced. If half-lives are available for the three phases, an overall half-life can be deduced essentially by weighting the rate constants (which are related to the reciprocal half-lives) in these partitioning proportions. This results in a distribution of persistence half-lives at 25°C.

A method was devised to express the temperature dependence of the partition coefficients using typical enthalpies of phase change. The substances were then run through the estimation method at 0°C to determine how the environmental distribution would change and the extent to which persistence would increase or decrease. The results are shown in Figures 1a and 1b which are plots of K_{AW} versus K_{OW} on logarithmic scales. The diagonals are lines of constant K_{OA}.

The points tend to move down and to the right towards the region where octanol dominates. Specifically, K_{AW} decreases by a factor of 7.6 and K_{OW} increases by a factor of 2.1, so K_{OA} increases by a factor of 16, i.e. 1.2 decades on the log plot. The implications are as follows:

- Half-lives in soils and vegetation become more important at low temperatures.
- Some volatile substances which tend to partition into air at warmer temperatures, migrate to condensed media at lower temperatures, i.e. they become more "multimedia" in character). Specifically, the number of chemicals in the > 99% air region to the upper left decreases from 23 to nine. The number in the > 99% octanol region to the lower right increases from 42 to 63. Clearly the multi-media character of these substances changes.
- There will be an increase in persistence because degradation rates are usually slower in condensed media than they are in the atmosphere.

The literature was searched for reasonable values of activation energies which would characterize the temperature dependence of the half-lives. The set of chemicals was then run for the partitioning data at 25°C and 0°C. The distributions of the resulting persistences are given in Figure 2. Clearly, the 25°C drop in temperature has a profound effect on persistence. The overall results suggest that estimates of persistence at the usual 25°C may be inappropriate for cold climate conditions because at (say) 0°C partitioning changes cause persistence to increase by a factor of about 2 and reaction rate changes cause a parallel increase of a factor of about 3, resulting in a total factor of 6. For other temperature changes ΔT the factors can be estimated approximately as:

$$6\ \Delta T/25$$

These increases are substantial and bring into question the entire procedure of using "cut-off" criteria for persistence at 25°C during international assessments. A general conclusion is that it becomes more important to determine degradation rates in soils at low temperatures.

Long-range transport

A reduction in temperature, as is experienced by a chemical moving to the Arctic, causes two changes in properties, which affect long-range transport. First, degradation rates are slowed, thus the chemical persists longer and has the potential to travel greater distances. This is particularly important for atmospheric reactions which are largely driven by hydroxyl radical concentrations, which in turn are controlled by solar insolation. The net effect is a decrease in the rate constant for reaction (k_R) and an increase in long-range transport distance.

The second effect is to modify partitioning from the vapour phase to "condensed" phases such as aerosol particles, soils, water and vegetation. A reduction in temperature causes the chemical to favour the condensed phase, e.g. the partition coefficient between air and water (K_{AW}) falls. Thus, there is a tendency for the chemical to be more rapidly removed from the atmosphere to terrestrial, aquatic and marine compartments, and to remain longer in these "condensed" phases.

This rate can be expressed as a deposition rate constant k_D which is a function of the partition coefficient. Further, the fraction of the chemical which remains permanently in the condensed phase F (which we have referred to as the "stickiness") is generally increased because evaporation is slowed, but degrading reactions in the condensed phase are also slowed, thus the chemical may persist longer and have an opportunity to evaporate. It is thus possible that in some cases F may decrease.

Combining these terms is best done with the relatively simple equation expressing the distance that half of the chemical will travel in air. A quarter will travel twice this distance etc.

$$\text{Long-range transport half distance} = 0.69\ \text{wind velocity}/(k_R + k_D F)$$

For example, if we take a wind velocity of 15 $km \cdot h^{-1}$, if k_R (reaction) is 0.01 h^{-1}, k_D is 0.02 h^{-1} (deposition) and F is 0.5 (half remains permanently on the surface and half reacts) the long-range transport distance is:

$$0.69 \times 15/(0.01 + 0.02 \times 0.5) = 517\ \text{km}$$

At low temperatures, k_R may fall to 0.001 h^{-1}, k_D may increase to 0.03 and F may fall to 0.3 because of slower degradation in soils. The resulting long-range transport distance will be a longer 1035 km. But if the decrease in k_R is only to 0.008 h^{-1} and F is unchanged ,the distance will be a shorter 450 km. Thus, it is clear that there are competing tendencies and whether long-range transport distance increases or decreases at lower temperatures depends on the interplay between values of k_D, k_R and F.

In this project, the scientific foundation of this effect has been established and the remaining future task is to apply these concepts to a variety of real chemicals in a manner comparable to that done for persistence. A full report on this issue is in preparation.

Discussion and Conclusions

International negotiations to identify and regulate POPs which are of concern in the Canadian North will inevitably involve assessment of the attributes of a large number of candidate substances with priority being assigned to those of most concern from the viewpoints of persistence, bioaccumulation, toxicity and potential for long-range transport. As a result of this study, it is now quantitatively demonstrated that a drop in temperature of say 25°C will have a profound effect on persistence. The factor by which the persistence will increase appears to be about 6, but chemical-to-chemical variation is expected, thus ranked lists of chemicals may change in order as temperatures change. Certainly, absolute values will increase considerably. This information should be of value to those undertaking international negotiations, especially when it is published in the refereed scientific literature, as is planned.

Similarly, we have evaluated the effect of a temperature drop on potential for long-range transport. This proves to be more complex than was first expected, but a sound scientific basis has been established for making assertions about the direction and magnitude of the change. Several complicating factors remain to be examined more fully. The role of snow as a modifying factor to POP transport is not well understood (Wania et al. 1999b). Likewise it is suspected that vegetation can play a considerable role by trapping POPs. Since much of the terrain below the Canadian tree line is forested, it is possible that these forests act as a "filter" or "barrier" to POP transport from south to north. Changes in land altitude as occurs in mountain ranges may also have a profound effect due to air mass cooling and enhanced snow fall. Elevated levels of POPs have been detected in snow and water at high altitudes (Blais et al. 1998).

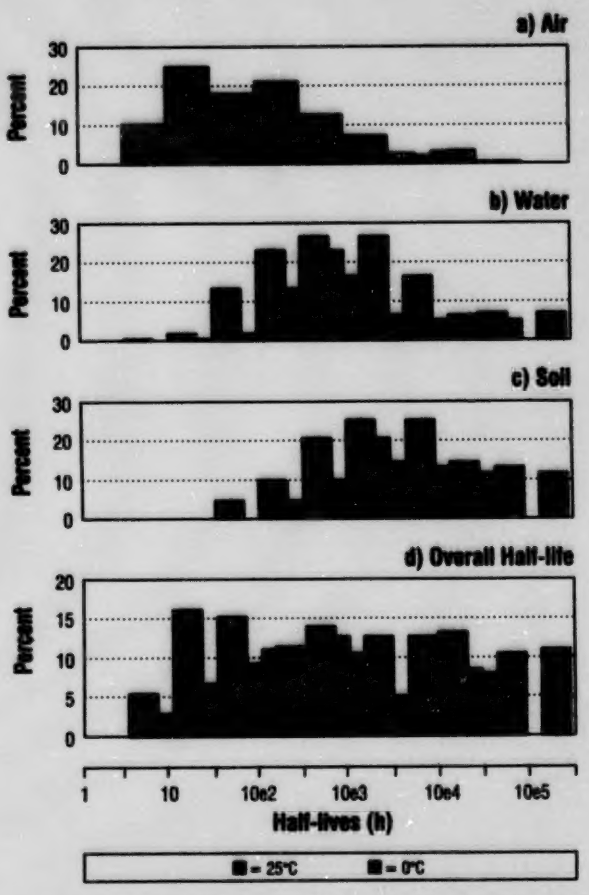
In our view, what is needed to assist in international negotiations is at least a four-part strategy, many of the aspects of which are already in place.

First is a comprehensive monitoring program to detect, quantify and report the presence and levels of POPs throughout the abiotic and biotic ecosystems of the Canadian North.

Second is a research program involving experimental and modelling studies to establish the inevitability that these POPs have migrated to the North and how they partition, persist and accumulate in that environment. Presumably not all chemicals can make the long journey and they will differ in persistence, thus a sound method of discriminating between chemicals or ranking them at both cold and temperate conditions is needed. The work described here on persistence and long-range transport is aimed at contributing to this effort.

Third is an attempt to prepare an internationally accepted global model of chemical fate which can be used to evaluate the fate of POPs in a manner similar to that done for γ-HCH (Wania et al. 1999a and Wania and MacKay 1999a). The model can only achieve international credibility if it is transparent, can be used by a variety of users internationally and can be shown to give results for a number of chemicals which are consistent with observation (Wania and Mackay 1999b). The work to make the Wania-Mackay model available to all users fulfills part of this objective. It is probable that other models will be developed, thus it will be important in the future to compare the findings of different models from different research groups.

Figure 2. Distributions of individual phase half-lives and overall half-life resulting from a change in temperature from 25°C (open bars) to 0°C (shaded bars)*



* The assumed enthalpies of phase change are air-water 55 kJ·mol^{-1}, octanol-water –20 kJ·mol^{-1} and octanol–air –75 kJ·mol^{-1}. Assumed activation energies are: air 14 kJ·mol^{-1}, all other media 47 kJ·mol^{-1}. No account is taken of variation in hydroxyl radical concentrations which would add another factor decrease in rate in air.

Finally, there is a need to inform the lay public as well as those responsible for chemical management about the status of research on contamination of northern ecosystems, including implications for human health. The "popular" article is designed to satisfy this need.

Work is continuing on all the topics addressed above in 2000-2001, specifically:

- further testing and possible modification of the global model
- application of the model to other substances, e.g. PCBs and DDT
- continued examination of persistence criteria
- continued examination of factors influencing long-range transport potential
- compilation of data and estimation methods necessary to assess chemical fate in cold climates.

A decision was made not to seek NCP funding for 2000-2001, but these aspects are being addressed using funding from other sources such as NSERC. It is likely that a funding request will be made for 2001-2002.

Expected Project Completion Date

The project in its present form is expected to be completed in July 2002.

Acknowledgments

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An Assessment of Sources of HCH Isomers to Wildlife and Humans in the Canadian Arctic

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Abstract

The objectives of this 1-year study were to examine sources, fate and bioaccumulation of HCH (hexachlorocyclohexane) isomers, especially β-HCH, from seawater and air to human milk using existing data from Northern Contaminants Program Phase I and other studies, to study possible isomerization of HCH isomers, particularly lindane to α- and β-HCH, and to prepare an exposure assessment of the elevated levels of β-HCH in humans in the Canadian Arctic. After compiling a large number of recent results for HCH isomers, we found a general increase in the proportion of β-HCH in ΣHCH with trophic level. The proportion of β-HCH in ΣHCH ranged from 0.03% in the benthic amphipod *Anonyx nugax* (where β-HCH was at detection limits) to 96% in human milk. The bioaccumulation factor for β-HCH was similar to that for α-HCH in benthic invertebrates and zooplankton but 10x higher in Arctic cod and seabirds. Similarly, biomagnification factors for β-HCH from Arctic cod to seabirds and from seals to polar bears were higher than for α-HCH and γ-HCH. This appears to reflect the much greater stability of β-HCH than the other HCH isomers although lower than biomagnification factors for PCB 153, a highly persistent PCB congener. Enantiomer ratios of α-HCH in mussels, char and seals were positively correlated with β-HCH/ΣHCH ratio indicating that β-HCH increased while α-HCH was being transformed. Degradation rates of β-HCH were high in polar bear liver microsomes suggesting rapid transformation of HCH isomers accumulated in the diet must be occurring. This study has shown that the elevated proportions and levels of β-HCH in human milk of Inuit in the Canadian Arctic are not due to elevated levels in diet although clearly some traditional foods of minor importance, such as polar bear meat or seabird eggs, will have high proportions. The recalcitrance of β-HCH and its long half-life in humans appear to be the best explanation for the observed levels.

Objectives

1. To examine sources, fate and bioaccumulation of HCH (hexachlorocyclohexane) isomers, especially β-HCH, from seawater and air to human milk using existing data from Northern Contaminants Program (NCP) Phase I and other studies.
2. To review the literature on possible isomerization of HCH isomers, particularly lindane to α-HCH and β-HCH.
3. To prepare an exposure assessment of the elevated levels of β-HCH in humans in the Canadian Arctic.

Introduction

Background

The NCP "Blueprint for Monitoring the Health of Arctic Peoples and Ecosystems and the Effectiveness of International Controls" recommended in Section 1.2 that priority should be given to the origin of α-HCH and β-HCH in the Arctic upper food chain biota. This 1-year project was designed to address that recommendation.

Literature review

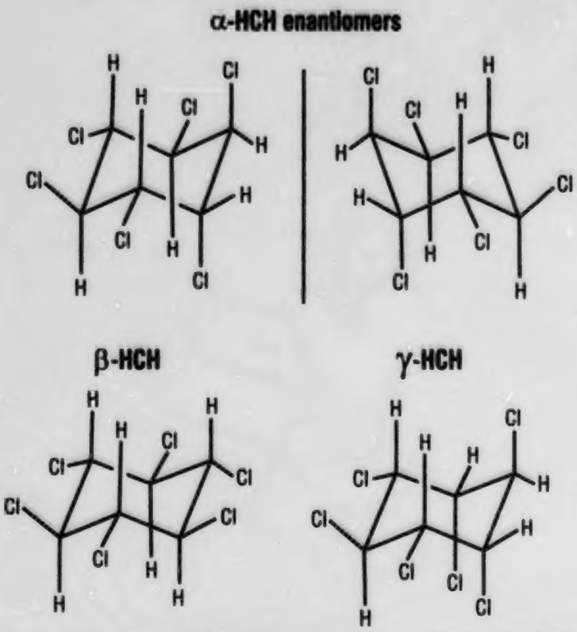
There have been two recent reviews of HCH toxicity, fate and environmental distribution (Willett et al. 1998, Walker et al. 1999). Both recommended further study of the extent and importance of isomerization of γ-HCH to α-HCH and especially the extent of bioaccumulation of β-HCH.

Synthesis and properties

HCH was first synthesized by Faraday in 1825 by reaction of benzene (bi-carburet of hydrogen) with chlorine in the presence of sunlight (Faraday 1825). The insecticidal properties of what Faraday called "benzene hexachloride" were first described in the 1940s and the active γ-isomer was named lindane after Van Linden, discoverer of the α- and γ-isomers (Brooks 1977). HCH is still occasionally called BHC in some listings of organochlorine chemicals. α-HCH is the only optically active isomer due to the unique positioning of chlorine substituents. The β- and γ-isomers do not have superimposable mirror images (Figure 1).

HCH has been manufactured as two predominant products: technical HCH (60% to 70% α-HCH) and lindane (γ-HCH). Technical HCH contains approximately equal amounts of β-HCH (5% to 14%) and γ-HCH (8% to 15%) along with other minor (ε, η and θ) isomers (Kutz et al. 1991). Only γ-HCH displays significant insecticidal properties.

Figure 1. Structures of HCH isomers including the optically active α-HCH isomer



α-HCH enantiomers

β-HCH

γ-HCH

The toxicology of α, β- and γ-HCH isomers has been studied extensively in mammals and to a lesser extent in fish and insects (Willett et al. 1998). The chronic effects of the β-HCH isomer are of particular interest because of its predominance among all HCH isomers in mammals, including humans, and its apparent long biological half-life. A half-life of 7.2 years has been estimated for β-HCH in humans (Jung et al. 1997) versus < 1 day for γ-HCH (Feldmann and Maibeck 1974). β-HCH has been shown to disrupt endocrine processes in mammals and fish at high doses. β-HCH administered in the diet of juvenile female rats at 50 $mg \cdot kg^{-1}$ caused significantly increased uterine weights (Loeber and van Velsen 1984) and induced vitellogenesis in male medaka fish exposed to 0.1-1 $mg \cdot L^{-1}$ in water (Wester and Canton 1986). However, mice fed up to 300 mg β-HCH·kg^{-1} diets did not show estrogen-like immunotoxic responses such as thymic atrophy and anovulation (Steinmetz et al. 1996). In summary, β-HCH has been described as producing estrogen-like effects through nonclassical estrogen-dependent mechanisms of action (Steinmetz et al. 1996).

The key physical properties that control the ultimate fate of HCH in the environment are high volatility, low Henry's Law Constant (K_H), a strong dependency of K_H on temperature (α-HCH partitions into water about seven times more strongly at 0°C than at 25°C [Kucklick et al.

1991]), low affinity for particles and resistance to degradation in cold water (Ngabe et al. 1993).

Historical and present uses

Technical HCH use began in 1943 and by the early 1990s 5.6 Mt of technical HCH and 0.7 Mt of lindane were produced globally (Li et al. 1998a, Li et al. 1998b). Technical HCH use was banned in most western countries and Japan in the 1970s but continued in India, Russia and China. China is reported to have been the major world producer accounting for about 4.5 Mt between 1945 and 1983. In 1983, China switched to lindane and Russia banned technical HCH in the late 1980s (Li et al. 1998a). In 1990, India banned the technical product for agricultural use but kept it for public health uses. Although HCH isomers were most heavily used by China and India, North America and Europe have also contributed significantly to global emissions. The banning of technical HCH in China in 1983 contributed to a dramatic downturn in air concentrations in the North American Arctic in the early 1980s (Li et al. 1998a). By 1991, only 24 000 tonnes were applied, most of it in India, although significant undocumented use may have also occurred in China (Li et al. 1998a). Lindane is still used worldwide, including in North America and Europe.

In Canada, lindane is used for seed treatment, especially in the prairies. A reduction in α-HCH concentrations was observed in water samples from rivers on the Canadian prairies over a 20-year period, however, concentrations of lindane have remained relatively constant and detection frequency has increased in rivers draining agricultural areas (Crosley et al. 1999). The proportion of lindane, of total HCH, in Lake Winnipeg sediment cores has increased in the past 15 years (Rawn et al. 2000). Elevated levels of lindane have been found in air above agricultural fields in Quebec (Poissant and Koprivnjak 1996).

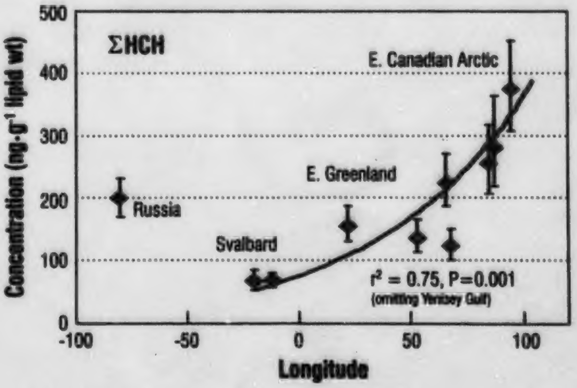
Ensuing reductions in HCH use by India and Russia (Iwata et al. 1993a, Li et al. 1998a) in the early 1990s have led to further atmospheric declines (Macdonald et al. 2000). The strong partitioning of HCH from air to water means that water phases ultimately become more important reservoirs than the atmosphere, containing > 90% of the inventory. The result of the dramatic decrease in atmospheric HCH concentration is that the surface water in some oceanic regions became undersaturated and therefore started to outgas (Jantunen and Bidleman 1996, Jantunen and Bidleman 1998).

Presence in the Arctic

HCHs are the most abundant organochlorine pesticides in the Arctic Ocean. HCHs evaporate quickly from temperate and tropical regions (Takeoka et al. 1991), disperse through the atmosphere and deposit through air-water gas exchange and precipitation. Partitioning of HCHs from air to water is favoured at lower temperatures (Kucklick et al. 1991) with the result that concentrations of HCHs in the Arctic Ocean are typically an order of magnitude or higher than those in tropical waters. Thus, HCHs provide a classic demonstration of the "cold condensation effect" (Wania and Mackay 1996). HCHs enter the Arctic Ocean via ocean currents on the Pacific side through the Bering Strait and on the Atlantic side through the Fram Strait and the Norwegian coastal current. Atmospheric processes also deliver HCHs by precipitation and air-water gas exchange. The distribution of HCHs in the Arctic Ocean is far from uniform. Spatial trends in surface water indicate that concentrations are highest in the Beaufort Sea and Canadian Archipelago, intermediate in the Bering-Chukchi seas and along a transect across the polar cap, and lowest in the Barents Sea and eastern Arctic Ocean (Macdonald et al. 2000, Harner et al. 1999). The elevated residues of HCHs in marine mammals of the Archipelago are likely due to drainage of water containing high concentrations of HCHs from the polar mixed layer (Barrie et al. 1997, Hargrave et al., 1997). Muir et al. (2000a) found highest levels of ΣHCH in ringed seals from the central Canadian Archipelago in comparison with results from Greenland, Svalbard and Russia (Figure 2).

Characteristic ratios of α-HCH/γ-HCH have been found in different water masses of the Bering-Chukchi seas (Rice and Shigaev 1997). This suggests regional inputs of HCH products having different isomer ratios, or selective breakdown of the isomers in different water masses. In support of the latter, enantioselective degradation of

Figure 2. Geographic trends of ΣHCH (means ± 95% confidence intervals) in ringed seals in the Arctic showing significantly higher levels in the Canadian Arctic*



* Source: Muir et al. 2000a

α-HCH has been found in polar waters, with a preference for depletion of the (+) enantiomer (mirror image isomer) in the western and eastern Arctic Ocean and the (–) enantiomer in the Bering-Chukchi seas (Harner et al. 1999, Jantunen and Bidleman 1996). Harner et al. (1999) estimated that breakdown of HCHs in the eastern Arctic Ocean takes place three to 10 times faster by microbial processes than hydrolysis.

Possible isomerization of HCH isomers

The ratio of α-HCH to γ-HCH in technical grade HCH varies between four and seven (Iwata et al. 1993b). The use of lindane lowers the ratio expected to be found in the environment. Concern about the potential for conversion of γ-HCH into other isomers, particularly α-HCH, comes in part from the observed high concentrations of the α isomer relative to the γ isomer in the northern hemisphere. However, air and water samples far from pesticide application sites have often contained ratios much higher than seven (Iwata et al. 1993b, Patton et al. 1989, Pacyna and Oehme 1988). Several hypotheses have been suggested to explain why so much of the HCH residue found in the environment is in the form of the α isomer.

One explanation is that γ-HCH is transformed by sunlight into α-HCH (more stable in sunlight) during long-range transport (Barrie et al. 1992, Pacyna and Oehme 1988, Oehme 1991). Laboratory experiments by Steinwandter (1976) and Hamada et al. (1981) found that α-HCH was the main decomposition product of crystalline γ-HCH exposed to UV irradiation, while the other isomers were quite stable under similar conditions. Malaiyandi and Shah (1984) also produced significant amounts of α-HCH from γ-HCH and from β-HCH, through aqueous photolysis using direct sunlight and ferrous salts. These experiments clearly indicate that isomerization is possible.

Oehme (1991) found that the α/γ ratio was lower in the winter and spring (when light and therefore photoisomerization rates would be lower) and higher in the summer and fall (when Arctic air masses are fairly isolated and photoisomerization rates would be higher), and suggested the seasonal fluctuations of the α/γ ratio was evidence for temperature-dependent photoisomerization. However, other factors such as the different rates of atmospheric volatilization and deposition of the isomers, as well as seasonal use of lindane or technical grade HCH, could also explain the fluctuations in the α/γ ratio (Halsall et al. 1998, Oehme et al. 1996, Hoff et al. 1992, Patton et al. 1989). Furthermore, a study of seasonal fluctutations of the α/γ ratio in Quebec (Poissant and Koprivnjak 1996) found no increase in α-HCH levels following applications of lindane in spring, suggesting either that photoisomerization is not the main process acting upon γ-HCH or that the sample sites were too close to application sites to allow time for significant photoisomerization to occur (photoisomerization presumably occurs during more long-range transport). Brubaker and Hites (1998) found an approximately 25% longer atmospheric lifetime for α-HCH than γ-HCH in air (120 versus 96 days) based on reaction with the hydroxyl radical in laboratory experiments. The higher proportions of α-HCH may therefore be explained by its longer half-life rather than isomerization.

Bio-isomerization in soil and sediments

There is evidence from laboratory studies that γ-HCH can be biotransformed into other isomers in soil or sediments. The orientation of Cl atoms of the γ isomer makes irreversible transformation into α-HCH the most likely form of isomerization (Newland et al. 1969, Buser and Müller 1995). Newland et al. (1969) found that 80% of the γ-HCH in a simulated lake impoundment was depleted within 3 months and that the main metabolites were α-HCH and δ-HCH. Benezet and Matsumura (1973) showed that γ-HCH could be transformed into α-HCH and several other compounds by a strain of bacteria *Pseudomonas putida*. Huhnerfuss et al. (1992) found that marine microorganisms in North Sea sediments could transform γ-HCH into essentially racemic (1:1 ratio) mixtures of both enantiomers of α-HCH, but did not specify the rate of transformation or the percentage of γ-HCH transformed. Recent quantitative studies have indicated that bio-isomerization is an insignificant part of the overall degradation of γ-HCH. Waliszewski (1993) found that field applications of lindane to soil produced only trace amounts of other HCH isomers. In their study of degradation of HCH isomers in anaerobic sewage sludge, Buser and Müller (1995) found that a very small percentage of γ-HCH could be transformed into α- or δ-HCH. Thus soils treated with lindane are not anticipated to be a major source of α-HCH. Recently, studies carried out on anaerobic and aerobic sediments showed bio-isomerization of α- → β-HCH, α- → γ-HCH and α- → δ-HCH. β-HCH was the most persistent isomer in their field environment due to bio-isomerization of α-HCH into β-HCH, as well as due to its greater persistence (Wu et al. 1997).

Biotransformation in biota

β-HCH is present at low levels in Arctic air (Halsall et al. 1998) and seawater (Jantunen and Bidleman unpublished data) and is found in increasing concentrations at higher trophic levels. The percentage of β-HCH of ΣHCH does not increase smoothly with trophic level. Much higher proportions of β-HCH are found in polar bear fat than in their prey species (Muir et al. 1988). Proportionally much higher β-HCH is found in human milk from northern Quebec (Muir and Dewailly, unpublished results 1991) than in marine mammal oils or Arctic char. The proportions of α-, β- and γ-HCH isomers also show large differences in the blubber of various marine mammals (Willett et al. 1998).

Specific accumulation of HCH isomers is also found in different tissues. In fur seals, β-HCH is the most abundant isomer in blubber and liver, but α-HCH is far more prevalent in brain (Mössner et al. 1992). Furthermore, enantiomeric analysis shows that residues of α-HCH in brain consist almost exclusively of the (+) enantiomer, suggesting enantioselective penetration through the blood-brain barrier (Mössner et al. 1992, Möller et al. 1994).

It has been suggested that the enantiomer ratios (ERs) of α-HCH could be used as a tool to determine the metabolic capacity of HCH for different species (Tanabe et al. 1996). The ER of α-HCH was inversely proportional to the β-HCH/ΣHCH ratio in small cetaceans. Low β-HCH/ΣHCH ratio indicates a high metabolic capacity since β-HCH is the most persistent HCH isomer in the animal body (Tanabe et al. 1996). The ER of α-HCH has been reported to be close to racemic in many fish species and blue mussel (Pfaffenberger et al. 1992, Wiberg et al. 1998a) while mammals in general preferentially accumulate (+) α-HCH (Mössner et al. 1992, Pfaffenberger et al. 1992, Wiberg et al. 1998a).

The ER of α-HCH has also proven to be higher in seal and polar bear livers than in their blubbers indicating that the higher enzyme activity of the liver may influence the ER (Mössner et al. 1992, Klobes et al. 1998 and Wiberg et al. 1998b). Klobes et al. (1998) and Norstrom et al (1988) also found that in adipose tissue of polar bear, the concentration of α- and β-HCH were 10× higher than the concentration of γ-HCH while the liver contained equal amounts of all three isomers. Furthermore, Klobes et al. (1998) found that the ER of α-HCH was close to racemic in the adipose tissue while the blubber contained significantly more of the (+) α-HCH.

Different ratios of β-HCH/α-HCH in polar bear liver versus adipose tissue could also be due to selective binding. In the case of polar bears, almost all oxygenated organochlorines, from dieldrin, to oxychlordane, have a significantly higher concentration in liver (Norstrom et al. 1988). This may have something to do with the high lipid content of stellate cells which are specialized in storing retinol esters. In all mammals, including humans, exposure to high levels of retinol esters in their diet induces proliferation of these cells. In addition to this, there is the binding of some chlorinated pesticides to cytochrome P450 proteins. Seabirds are a different matter. Birds generally seem to accumulate β-HCH. Whether this is bio-isomerization or selective bioaccumulation is not known.

The ER of α-HCH increases up the Arctic food chain from polar cod to seal to polar bear (Wiberg et al. 1998a). These results suggest that α-HCH is metabolized to an increasing extent and this is further supported by the lack of biomagnification of α-HCH in this food chain. All these reports suggest that α-HCH is degraded or transformed by mammals. However, little or no attention has been given to the products of this process. Whether α-HCH is bio-isomerized to β-HCH or dechlorinated to β-PCCH in mammals remains unclear. Iwata et al. (1998) found a significant relationship between the ER of α-HCH in the fat of fur seals and their feeding habit. ER values in the seals ranged from approximately1.2 to 2.2 (selective accumulation of the (+) enantiomer). Higher ERs were found in seals whose diet consisted mainly of fish compared to those whose stomach contents were mainly squid. Tanabe et al. (1996) examined ER and bioaccumulation relationships for cetaceans collected in the Bering Sea, North Pacific and Sea of Japan. ER values in blubber ranged from approximately1.5 to 2.8. A significant (negative) correlation was found between ER and the ratio of β-HCH/ΣHCH. Differences in ERs of α-HCH were also seen for diseased and healthy eider ducks (Pfaffenberger et al., 1992). The above studies indicate that accumulation of HCH isomers and enantiomers is influenced by trophic level, feeding habit, selective metabolism, membrane penetration and health status of the animal.

Activities in 1999-2000

To meet the project objectives, information from several independent projects led by the project leaders was assembled.

First, a review and synthesis of existing data for HCH isomers in air, water and biota in the Canadian Arctic was conducted, with emphasis on samples from the eastern Arctic where high levels of β-HCH have been reported in human blood and milk. Biomagnification of β-HCH was compared with the recalcitrant PCB congener CB153 and the fraction of β-HCH in various fish and marine vertebrates including ringed seals, polar bears and marine birds, was determined using results from existing samples.

Second, new results for β-HCH, as well as other HCH isomers, in seawater, sediment and food web components from the North Water study (Moisey et al. 2000) were examined.

Third, ERs of α-HCH (the chiral isomer) were examined in existing extracts of Arctic char, blue mussels and ringed seal samples from Northern Quebec to discern the extent of transformation of this congener.

Fourth, the bio-isomerization of α- and γ-HCH isomers to β-HCH was examined directly using polar bear liver microsomes. Two attempts were made to get fresh ringed seal liver for preparation of microsomes in May 1999 at Holman Island and in December 1999 at Clyde River. However, both attempts failed because livers could not be kept preserved at below –80°C during transport.

Chiral gas chromatography analysis

The enantioselective separation of α-HCH in the biological samples was performed on an HP6890 gas chromatograph using a chiral β-DEX with detection using electron capture negative ion mass spectrometry (HP5973MSD).

Bio-isomerization study

Individual HCH congeners were incubated with individual or pooled liver microsomes from polar bears in the laboratory of S. Bandiera at the University of British Columbia. After extraction and clean up, the amount of HCH remaining was determined by gas chromatography with electron capture detector (GC-ECD) or gas chromatography mass spectrometry (GC-MS). Initial experiments were conducted with pooled polar liver microsomes to optimize assay conditions. Microsomal fractions were originally prepared by differential centrifugation at 5°C (Thomas et al. 1983). Incubations with HCH isomers were carried out at 37°C (0 to 30 min) in 1M Hepes Buffer, pH 7.8, with 5mM $MgCl_2$. The incubation buffer contained NADPH (1 to 2mM). Hexane was added to stop the reaction and extract the products. Products of the reaction were analyzed by GC with electron capture detection.

Results

β-HCH and other HCH isomers in the Arctic marine food web

Results for β-HCH, other HCH isomers and the recalcitrant PCB congener 153 (2,4,5,2',4',5'-hexachlorobiphenyl) were assembled for seawater, sediment and 26 Arctic species ranging from benthic invertebrates to polar bears. Results for three human milk samples from northern Quebec provided by É. Dewailly to D. Muir were also included. Except for benthic invertebrates and seabirds, which were from the North Water study in North Baffin Bay, we used data from at least two locations. For marine mammals and Arctic char we used mean concentrations from male and females from at least two sites in the eastern Arctic including Nunavik and Hudson Bay (Table 1).

There is a general increase in the proportion of β-HCH in ΣHCH with trophic level (Figure 3). The proportion of β-HCH in ΣHCH ranged from 0.03% in the benthic amphipod *Anonyx nugax* (where β-HCH was at detection limits) to 96% in human milk (Table 1, Figure 3). These are average proportions for selected species at these locations; actual range of the % β-HCH in ΣHCH would be higher. Marine mammals and birds had high proportions of β-HCH with the lowest in ringed seals (15% to 27%) and beluga (20% to 23%) and highest in polar bear (41% to 92%) and seabirds (62% to 96%). Proportions of β-HCH in seawater were slightly lower than the fraction in technical HCH (3% to 5%). Proportions in sediment were higher (18%) than in seawater (2.6% to 5.1%) and similar in the surface slice of a core from central Hudson Bay and in surficial sediments from 14 locations in north Baffin Bay (Table 1 and Figure 3). There was considerable regional variation in the proportions of β-HCH in biota. In ringed seals, seabirds, beluga and Arctic char the proportions of β-HCH in ΣHCH did not vary with sex and/or age of the animals. Thus the regional differences (e.g. between ringed seals in north Baffin Bay and Labrador) (Table 1) may reflect actual differences in the food web and in seawater. β-HCH in ΣHCH was two-fold higher in ringed seals from north Baffin Bay (based on two separate studies) than in west Hudson Bay, and coincidentally also two-fold higher compared with ringed seals in Labrador and Northern Quebec. However, this was not the case for β-HCH in ΣHCH in beluga whales (Table 1).

The bioaccumulation factors (BAF = concentration in fat divided by concentration in seawater) for β-HCH were similar to those for α-HCH in benthic invertebrates and zooplankton but 10× and 100× higher in Arctic cod and seabirds, respectively (Table 2). Similarly biomagnification factors (BMF = concentration in fat of predator divided by concentration in fat of prey species) for β-HCH were higher than for α-HCH and γ-HCH, with the exception of Arctic cod to ringed seals. This appears to reflect the much greater stability of β-HCH than the other HCH isomers although, with the exception of zooplankton to Arctic cod, BMFs were lower than for CB153.

The ratio of β-HCH to CB153 was examined in the same samples (Table 1) to assess the relative bioaccumulation potential of the β isomer. Ratios to CB153 as well as

Table 1. Compilation of recent data for % β-HCH in ΣHCH and β-HCH/CB153 ratio in seawater, sediments and marine biota from the eastern Canadian Arctic

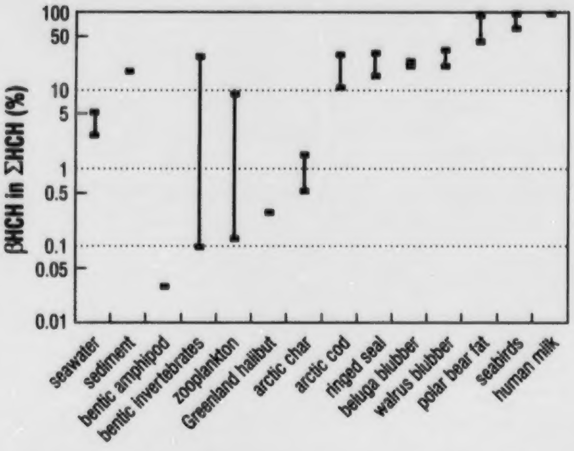
Sample type	Species	Location	n*	% β-HCH of ΣHCH	β-HCH/ CB153	Reference
seawater	seawater	West Hudson Bay	19	2.6	6.7	Moisey et al. 2000
seawater	seawater	Northern Baffin Bay	3	5.1	10.4	Muir and Lockhart, unpublished
sediment	sediment	Northern Baffin Bay	14	17.6	1.0	Moisey et al. 2000
sediment	sediment	Central Hudson Bay	1	17.6	0.23	Muir and Lockhart, unpublished
benthic invertebrate	*G. arcticus*	Northern Baffin Bay	3	0.25	0.02	Moisey et al. 2000
benthic invertebrate	*C. crispatus*	Northern Baffin Bay	2	23.5	0.08	Moisey et al. 2000
benthic invertebrate	*Y. thraciceformis*	Northern Baffin Bay	2	27.3	0.04	Moisey et al. 2000
benthic invertebrate	blue mussel	Hudson Strait	2	0.20	0.16	Muir et al. 2000b
benthic invertebrate	blue mussel	Ungava Bay	2	0.10	0.04	Muir et al. 2000b
benthic amphipod	*A. nugax*	Northern Baffin Bay	5	0.03	0.002	Moisey et al. 2000
zooplankton	Zooplankton	West Hudson Bay	3	24.5	0.26	Muir and Lockhart, unpublished
zooplankton	*M. occulata*	Northern Baffin Bay	7	8.8	0.13	Moisey et al. 2000
zooplankton	*Sagitta* spp.	Northern Baffin Bay	6	8.1	0.17	Moisey et al. 2000
zooplankton	*E. glacialis*	Northern Baffin Bay	3	0.40	0.03	Moisey et al. 2000
zooplankton	*C. hyperboreus*	Northern Baffin Bay	20	0.33	0.03	Moisey et al. 2000
zooplankton	*M. longa*	Northern Baffin Bay	3	0.27	0.006	Moisey et al. 2000
zooplankton	*T. libellula*	Northern Baffin Bay	4	0.12	0.005	Moisey et al. 2000
pelagic fish	Arctic cod	Northern Baffin Bay	8	30.0	1.11	Moisey et al. 2000
pelagic fish	Arctic cod	Lancaster Sound	3	10.6	0.06	Muir and Lockhart 1994
anadromous fish	Arctic Char	Ungava Bay	13	1.47	0.35	Muir et al. 2000b
anadromous fish	Arctic Char	Hudson Strait	6	0.50	0.12	Muir et al. 2000b
desmersal fish	Greenland halibut liver	Cumberland Sound	10	2.75	0.19	Muir and Lockhart 1996
desmersal fish	Greenland halibut muscle	Cumberland Sound	10	0.26	0.02	Muir and Lockhart 1996
walrus blubber	walrus blubber	Northern Baffin Bay	16	32.6	1.24	Muir et al. 2000c
walrus blubber	walrus blubber	Foxe Basin	24	20.2	0.67	Muir et al. 1995
beluga blubber	beluga blubber	South Hudson Bay	17	22.5	0.20	Muir, 1996
beluga blubber	beluga blubber	Hudson Strait	11	23.3	0.33	Muir, 1996
beluga blubber	beluga blubber	Northern Baffin Bay	24	19.6	0.16	Muir, 1996
seabirds	dovekie	Northern Baffin Bay	8	62.4	0.68	Moisey et al. 2000
seabirds	thickbilled murre	Northern Baffin Bay	9	68.3	0.22	Moisey et al. 2000
seabirds	black guillemot	Northern Baffin Bay	7	69.8	0.45	Moisey et al. 2000
seabirds	black legged kittiwake	Northern Baffin Bay	8	76.1	0.03	Moisey et al. 2000
seabirds	glaucous gull	Northern Baffin Bay	10	95.8	0.09	Moisey et al. 2000
seabirds	ivory gull	Northern Baffin Bay	4	89.2	0.03	Moisey et al. 2000
seabirds	northern fulmar	Northern Baffin Bay	10	63.3	0.01	Moisey et al. 2000
ringed seal	ringed seal	Northern Baffin Bay	57	29.1	0.21	Moisey et al. 2000
ringed seal	ringed seal	Northern Baffin Bay	8	27.0	0.14	Muir et al. 2000a
ringed seal	ringed seal	Hudson Strait	24	15.7	0.10	Muir et al. 2000b
ringed seal	ringed seal	Labrador coast	19	14.9	0.11	Muir et al. 2000b

Table 1. Compilation of recent data for % β-HCH in ΣHCH and β-HCH/CB153 ratio in seawater, sediments and marine biota from the eastern Canadian Arctic

Sample type	Species	Location	n*	% β-HCH of ΣHCH	β-HCH/ CB153	Reference
polar bear fat	polar bear fat	Admiralty Inlet	10	92.5	0.071	Norstrom et al. 1998
polar bear fat	polar bear fat	Resolute	16	41.1	0.042	Norstrom et al. 1998
human milk	human milk	Northern Quebec	3	96.1	0.31	Dewailly et al. 1993

* benthic invertebrates and zooplankton samples represent pooled whole organism samples

Figure 3. Percent of β-HCH in ΣHCH in a range of samples from the eastern Canadian Arctic*



* Vertical bars represent range of mean values. See Table 1 for sample list. Note that the vertical axis uses a log scale because of the wide range of values.

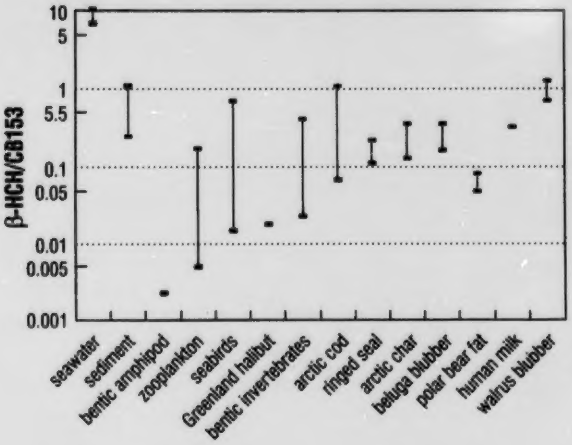
relative ratios (R_{rel}) (Compound X/CB153 in predator divided by Compound X/CB153 in prey) are often used to assess relative biotransformation (Boon et al. 1994) because the 2,4,5,2',4',5'-substituted PCB congener is known to be recalcitrant. β-HCH/CB153 ratios ranged from 0.002 to 10.4 (Table 1, Figure 4). Highest ratios were in seawater, reflecting relatively high levels of β-HCH compared to the much more hydrophobic CB153. In biota, a general increase in the β-HCH/CB153 ratio was seen from benthic amphipods to fish and mammals. However, a wide range of β-HCH/CB153 ratios were observed among species of seabirds, zooplankton, benthic invertebrates and Arctic cod (Figure 4). In general, this wide range reflects species differences in the ability to transform β-HCH as well as differences in feeding behaviour and trophic level of the seabirds.

R_{rel} values for seabirds/cod, ringed seals/cod and polar bears/seals were 0.36, 0.23 and 0.4, respectively, indicating that β-HCH is transformed relative to CB153. An R_{rel} value of 1 indicates no transformation from prey to predator. However, β-HCH has higher R_{rel} values than

Table 2. Average bioaccumulation factors (concentration in fat/concentration in water) and biomagnification factors (concentration in fat of predator/concentration in prey) for HCH isomers and PCB 153 in the marine food web, using results from Table 1

	α-HCH	β-HCH	γ-HCH	CB153
Bioaccumulation factors				
Benthic invertebrates	2.3×10^4	2.6×10^4	3.4×10^4	0.19×10^7
Zooplankton	1.5×10^4	1.2×10^4	4.8×10^4	0.14×10^7
Arctic cod	2.8×10^4	27.8×10^4	7.2×10^4	0.70×10^7
Seabirds	2.4×10^4	259×10^4	3.4×10^4	29×10^7
Ringed seal	7.2×10^4	46.5×10^4	2.7×10^4	2.8×10^7
Polar bear	8.0×10^4	289×10^4	0.5×10^4	44.8×10^7
Biomagnification factors				
Zooplankton to arctic cod	1.9	24.1	1.5	4.9
Arctic cod to ringed seals	2.6	1.7	0.4	4.1
Arctic cod to seabirds	0.9	9.3	0.5	41.7
Ringed seals to polar bear	1.1	6.2	0.2	15.8

Figure 4. Ratio of β-HCH to PCB 153 in a range of samples from the eastern Canadian Arctic*



* Vertical bars represent range of mean values. See Table 1 for sample list. Note that the vertical axis uses a log scale because of the wide range of values.

many PCB congeners with vicinal H in the meta and para positions (Boon et al. 1994, Muir et al. 1995, Fisk et al. 2000).

Enantiomer ratios of α-HCH

ERs of α-HCH in mussels, Arctic char and ringed seals were determined in 43 samples from northern Quebec and Labrador (Table 3). Lowest ER values (0.54 to 0.74) were found in mussels. These values are lower than observed for benthic invertebrates in the North Water study (Moisey et al. 2000). The lower ERs for mussels than for char or seals was surprising but may be a reflection of microbial degradation occurring on particles in the water column and in surface sediments. Slightly more non-racemic ERs for α-HCH were found on particles in seawater from the Chukchi Sea compared with the dissolved phase (Jantunen and Bidleman 1998). Therefore, our results for mussels may be explained by filtering of particles with α-HCH lower ERs than seawater. ERs of α-HCH declined from west to east in the Arctic Ocean and were < 1 in waters of Lancaster Sound (Falconer et al. 1995). Our results for mussels in Ungava Bay and Hudson Strait are consistent with the geographical trend of lower ERs of α-HCH in Arctic waters influenced by the Atlantic Ocean.

The ERs and enantiomeric fractions (EFs) in mussels, char and seals were positively correlated with β-HCH/ΣHCH ratio (ERs $r^2 = 0.16$, $p = 0.007$, $n = 43$; EFs $r^2 = 0.18$, $p = 0.004$). Tanabe et al. (1996) found the ER of α-HCH was inversely proportional to the β-HCH/ΣHCH ratio in small cetaceans and Moisey et al. (2000) also observed this correlation in the North Water study food web. This reflects the fact that the ER is a measure of the degree of biotransformation of α-HCH and that β-HCH is less readily transformed.

No significant differences (t-test, $p > 0.05$) were found for ERs of α-HCH between male and female char. There were significantly lower ERs in female compared to male ringed seals (Table 3). Karlsson et al. (2000) have previously shown that ERs for some chlordane components are gender specific in Atlantic cod (*Gadus morhua*). ERs for landlocked char were significantly higher (t-test, $p = 0.002$) than for sea-run char. This may reflect different sources of α-HCH for the landlocked fish. Helm et al. (2000) found ERs of α-HCH in water from three lakes on Cornwallis Island ranged from 0.71 to 0.74 compared with ER of approximately 1 in snow in the lake watersheds. ERs were significantly correlated with water temperature in Amituk Lake suggesting that rising temperatures following snow melt increased microbial degradation rates. The higher ERs in landlocked char therefore may be due to uptake of non-racemic HCH in the freshwater environment. By contrast sea-run char which feed mainly in the marine environment will reflect the near racemic ERs (0.93) observed in seawater in the Canadian Arctic (Falconer et al. 1995).

Table 3. α-HCH enantiomer ratios in mussels, arctic char muscle and ringed seal blubber samples from Northern Quebec (Ungava and Hudson Strait area) and Labrador (Nain)

Species	Sex	n	Mean	Minimum	Maximum
mussels (*Mytilus edulis*)	–	8	0.645	0.542	0.743
sea-run char	F	8	0.962	0.852	1.045
	M	15	0.907	0.857	0.992
landlocked char	F	3	1.139	1.072	1.206
	M	3	1.005	0.982	1.050
ringed seals	F	4	0.927	0.869	0.969
	M	5	1.074	0.933	1.416

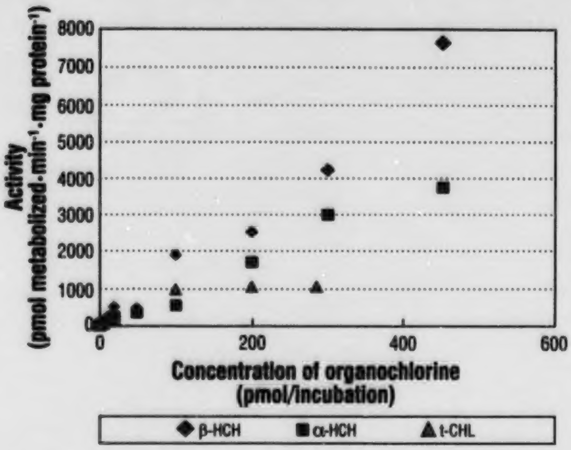
Bio-isomerization study

HCH isomers were incubated with polar bear liver microsomes to test for metabolism and possible bio-isomerization of α-HCH to β-HCH. Test assay conditions were selected to ensure that HCH metabolism was linear with respect to both incubation time and microsomal protein concentration and to select those substrate (HCH) and cofactor (NADPH) concentrations that were saturating. The results show that saturation with initial HCH concentrations up to 22.6 mM was not achieved for α-HCH or β-HCH although it was achieved for *trans*-chlordane, which was used as a reference compound (Figure 5). β-HCH was metabolized more efficiently by polar bear liver microsomes than α-HCH (indicated by the steeper slope of the β-HCH data points in Figure 5) and both HCH isomers are metabolized more efficiently than *trans*-chlordane. The results indicate that α- and β-HCH are rapidly metabolized by polar bear liver, and in fact are metabolized more readily than chlordane. Extracts were also analyzed for enantiomers of α-HCH and *trans*-chlordane. ERs were approximately 1 for both compounds in all samples tested. This suggests that polar bears do not degrade HCH isomers via an enantio-specific pathway. Further work is planned to examine the degradation rates and enantiomers in liver microsomes from 14 individual bears.

Summary and Preliminary Assessment

This study has developed new information on levels and bioaccumulation of β-HCH in the Canadian Arctic marine environment. The general increase in the proportion of β-HCH in ΣHCH and of the β-HCH/CB153 ratio with trophic level in the marine food web (except for seawater and sediment) provides evidence that it is simply more recalcitrant than the other two HCH isomers. The greater recalcitrance of β-HCH is also evident from much higher BAFs and BMFs of this isomer compared with other isomers. However, the β-HCH/CB153 relative ratios < 1 from prey to predator suggest β-HCH is metabolized in the marine food web relative to CB153, but is more recalcitrant than some lower chlorinated and 3,4-vicinal unsubstituted PCBs. Also, the microsomal degradation rates of β-HCH were high in polar bear microsomes suggesting rapid transformation of HCH isomers accumulated in the diet must be occurring.

Figure 5. Plot of rate of degradation (picomoles per minute per mg protein) at various concentrations of each HCH isomer and *trans*-chlordane incubated with polar bear liver microsomes*



* Steeper slope of β-HCH, degradation rate versus concentration, compared with α-HCH suggests more rapid degradation of β-HCH isomer.

The prevailing high proportions and levels of β-HCH in polar bear seem surprisingly high given the high microsomal transformation rates. The high levels of β-HCH require high proportions in their ringed seal diet, which is not the case. Ringed seals show much lower β-HCH/ΣHCH ratios than other top predators. Similarly, seabirds have predominantly β-HCH, which cannot be explained from levels in their diet, i.e. levels in fish or invertebrates. The high proportions of β-HCH in human milk could be a reflection of consumption of marine mammal and seabird meats and fat. However high proportions of β-HCH are found in all humans, not just those on a marine diet. The best explanation for elevated proportions of β-HCH in polar bears and seabirds, as well as humans, is therefore its greater recalcitrance than the other isomers. However, given the results for microsomal degradation in polar bears, the possibility of bio-isomerization cannot be completely ruled out at this time.

This compilation of β-HCH data also serves to show that elevated proportions and levels of β-HCH in human milk of Inuit in the Canadian Arctic are not due to elevated levels in diet although clearly some traditional foods of minor importance, such as polar bear meat or seabird eggs, will have high proportions. The recalcitrance of β-HCH and its long half-life in humans appear to be the best explanation for the observed levels.

Project Completion Date

March 31, 2000

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Atmospheric Mercury Transport, Oxidation and Fallout in Northern Quebec (Nunavik): an Important Potential Route of Contamination

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Abstract

Mercury is one of the most toxic contaminants in the environment. It is of concern in the North because environmental mercury levels there are elevated due to long-range transport as well as local emissions. Moreover, mercury may be chemically transformed and it bioaccumulates in the Arctic food chains. This atmospheric mercury project in Kuujjuarapik is in its first stage of development and is part of one of the first studies to conduct atmospheric chemistry measurements in northern Quebec. Important findings were made within this first year, 1999-2000. Background mercury in air in Kuujjuarapik is greater than at other remote locations in Canada; however, mercury in precipitation seems to be low, with a south to north decreasing concentration gradient. Mercury depletion events are extended over the whole Arctic area in the springtime. Fresh mercury deposition is volatilized further in the days following a depletion event, and mercury depletion seemed to be connected with bromine monoxide episodes.

Objectives

1. To measure total gaseous mercury (Hg) concentrations in air at Kuujjuarapik (Hudson Bay, 77°75′ W 55°28′ N) and to study their relationship with other locations in Canada, e.g. Canadian Atmospheric Mercury Network (CAMNet);
2. To study Hg oxidation processes by measuring ozone, chlorine (Cl) and bromine (Br) radicals in the air;
3. To measure total Hg concentrations in precipitation (as well as major ions, such as Cl^- and Br^-) in precipitation at Kuujjuarapik and to study their relationships with long-range transport and Hg fallout;

4. To measure Hg surface-air fluxes, especially during springtime;
5. To measure Hg concentrations in various abiotic components (soil, water, snow, etc.), over space and time in Nunavik and to study their potential connection with atmospheric Hg fallout.

Introduction

Hg is one of the most toxic chemicals in the environment. Its presence can be environmentally significant even in small quantities. Methylmercury (CH_3Hg^+), the most toxic form, can be bioconcentrate in the food chain on the order of 10^6 to 10^7.

The global Hg cycle involves several biogeochemical transformations between inorganic and organic Hg species. Accumulation of Hg by organisms is strongly dependent on the Hg cycle. In turn, the global cycle depends on the chemical form, subsequent chemical transformations and interactions with organisms. The ecosystem cycle of Hg between the aquatic (Poissant and Casimir 1998, Poissant et al. 2000, Amyot et al. 2000, Poissant et al. 1999) and atmospheric reservoirs is achieved through complex chemical (Poissant 1997) and biochemical reactions.

In northern Canada, Hg cycling is an important issue since environmental Hg levels are enhanced by anthropogenic activities and due, to a certain extent, to long-range transport. Also, it has been recently suggested that atmospheric Hg fallout can be amplified during polar sunrise (Schroeder et al. 1998). Although the mechanisms are still enigmatic, Br radicals have been pointed out as a potential oxidation species. Indeed, reactive Br species contribute significantly to the destruction of ozone in the polar stratosphere as well as the underlying troposphere (Barrie et al. 1988). Observation of bromine monoxide (BrO) (and chlorine monoxide [ClO], which is less important) at ground level during these ozone depletion events has revealed halogen oxide mixing ratios of up to 30 parts per trillion – sufficient to destroy 30 to 40 parts per billion of ozone typically present in the boundary layer within 1 to 2 days. Satellite observations show that tropospheric air masses enriched in BrO are always situated close to sea ice and typically extend over areas of about 300 to 2000 km (Wagner and Platt 1998). The BrO abundance remains enhanced for periods of 1 to 3 days, especially in springtime during polar sunrise.

Atmospheric Hg transport, oxidation and fallout in northern Quebec (Nunavik) might be an important potential route of contamination to the northern environment. The extent of atmospheric transport and deposition, however, has not yet been assessed and measured for Hg. The impact of Hg in the Arctic environment is evident. Where is this Hg coming from? How is it circulated and deposited into the Arctic region? These questions are of concern and very important in the Arctic as well as in a global perspective.

Activities in 1999-2000

Various activities were done in the first year of this new NCP research project. First of all, the monitoring research station was established in a safe location with available meteorological facilities located close by. The site is located in Kuujjuarapik, Nunavik, at the Centre d'étude nordique (CEN), Laval University. Meteorological data are available from CEN and Navigation Canada. Total gaseous Hg (TGM) and ozone instruments were installed by mid-August, and Hg and ozone data (TGM at 15-minute intervals, O_3 at 5-minute intervals) are collected daily through telemetry (Poissant et al. 1996). TGM measurements were audited (quality controlled) at the site during a national audit conducted by CAMNet in mid-November.

A cost effective and high quality monthly sampling protocol for Hg and major ions was elaborated and applied. Precipitation samples were collected since September, 1999. Precipitation samples are analyzed at IRNS-EAU (Dr. M. Amyot) for Hg, bromide and chloride . In spring 2000, Hg snow surveys were done in three locations in northern Quebec (Kujjuarapik, Kuujjuaq and Salluit). Sample analysis is ongoing. Also, snow-air Hg gas exchange flux experiments were conducted. These are the first Hg flux measurements conducted in the Arctic. Measurements of reactive gaseous Hg (RGM), TGM, ozone, TGM flux and Hg in snow were done simultaneously.

Daily satellite data for BrO concentration in the Arctic domain were collected by the Global Ozone Monitoring Experiment (Global Ozone Monitoring Experiment 2000).

Results and Discussion

TGM and ozone preliminary results

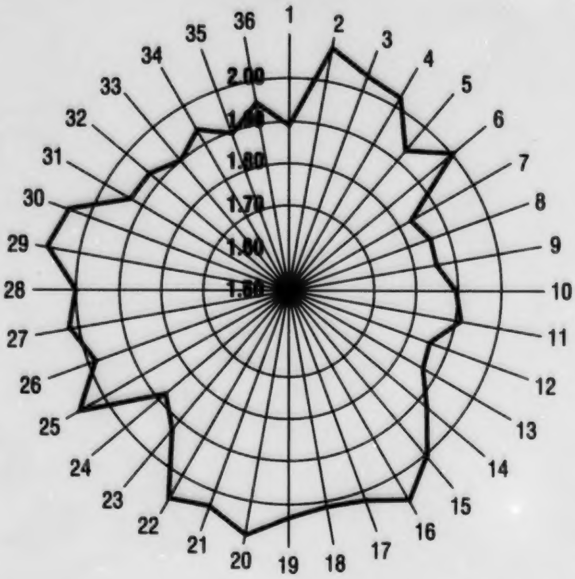
Table 1 presents a statistical summary of TGM and ozone measurements at Kuujjuarapik from August 1999 to March 2000. TGM results show higher concentrations (with median of about 1.98 ng·m^{-3}) than other sites located along the St. Lawrence River (Poissant 2000) and at Alert (Schroeder, personal communication). The average concentrations across CAMNet were approximately 1.6 ng·m^{-3} in 1998 (Kellerhals et al. 2000).

Table 1. Statistical summary of TGM and ozone in Kuujjuarapik, August 1999-March 2000

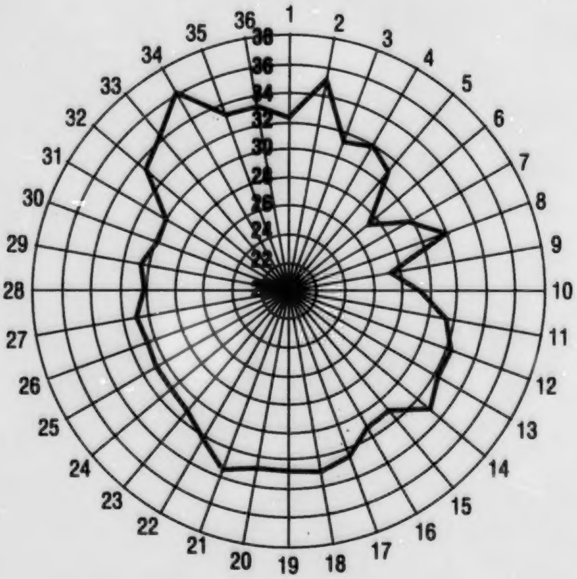
	TGM (ng·m^{-3})	Ozone (ppbv)
Median	1.98	32.05
Minimum	0.14	0.09
Maximum	8.96	52.12
Number	19 836	53 939

Figure 1. TGM and ozone roses in Kuujjuarapik, August 1999-March 2000

TGM rose (ng·m^{-3})
(Kuujjuarapik)



Ozone rose (ppbv)
(Kuujjuarapik)



Local sources of TGM are suspected. One potential local source would be the power plant in Kuujjuarapik. About 2 million litres of diesel are used by the power plant annually. Hg can be emitted from diesel burning (Poissant et al. 2000). The TGM related to this power plant source would involve large concentrations of RGM and greater Hg deposition in the vicinity. Since fishing, hunting and consumption of traditional foods occurs in the communities, local sources of Hg contamination would be an important potential source of contamination to the local environment and people.

Figure 1 gives the median TGM and ozone roses at Kuujjuarapik. The worst TGM wind sectors were from south to west (median 2.08 ng·m^{-3}) and the cleanest were from south-east to north-east (median 1.83 ng·m^{-3}). The minimum TGM and ozone concentrations were recorded as 0.14 ng·m^{-3} and 0.09 ppbv, respectively. Episodic low TGM and ozone concentrations were measured by mid-February, with maximum depletion events (Hg and ozone) measured by mid-March. Long-range transport as well as regional sources would impact the air quality since the background levels measured were still high in Kuujjuarapik. Further study using back trajectories will be done to find out the potential Hg source contributions (Poissant 1999).

Hg in precipitation

Hg in precipitation and some major ions (as complementary parameters) were measured on a monthly basis (Table 2). The median Hg concentration was 0.87 ng·L^{-1}. This median concentration is lower than the median concentrations observed along the St. Lawrence River (e.g. Saint-Anicet, 9.76 ng·L^{-1}; Mingan, 5.76 ng·L^{-1}) (Poissant and Pilote 1998). A south to north decreasing concentration gradient for Hg in precipitation is observed.

Snow-air Hg flux

A snow-air Hg flux campaign was conducted in the early spring 2000 with a complete survey of Hg concentration in snow, RGM, TGM and TGM flux. Figure 2 presents time series results for April 14-19, 2000. By April 16, 2000, a TGM and ozone depletion episode was observed during the TGM snow-air exchange experiments. TGM and ozone dropped close to zero during the night. Hg deposition was recorded during that depletion episode. Interestingly, TGM fluxes before the depletion event were about 0 to 0.5 ng·m^{-2}·h^{-1}, whereas TGM fluxes increased by

Table 2. Median concentrations of Hg and major ions in precipitation measured in Kuujjuarapik, September 1999-February 2000

	F^- ($mg\cdot L^{-1}$)	Cl^- ($mg\cdot L^{-1}$)	Br^- ($mg\cdot L^{-1}$)	NO_3^- ($mg\cdot L^{-1}$)	PO_4^{3-} ($mg\cdot L^{-1}$)	SO_4^{2-} ($mg\cdot L^{-1}$)	Hg ($ng\cdot L^{-1}$)	pH
Median	0.02	5.35	0.03	0.8	0	1.31	0.87	4.6
Number	6	6	6	6	5	5	6	3

about four times on the following days. This suggests that most of the fresh TGM deposition would be re-volatilized in the days following a depletion episode.

TGM and ozone depletion

As discussed above, TGM and ozone depletion events started by mid-February. TGM and ozone depletion was observed on April 16, 2000, but not on April 18, 2000, during the intensive snow-air exchange flux experiment. Those periods were compared to the Global Ozone Monitoring Experiment satellite observations for BrO concentrations (Figure 3). The depletion event on April 16 occurred during a large BrO cloud with extension over Kuujjuarapik, whereas this cloud was not extended as much during the April 18, 2000, depletion event. Further study will be done on the relationship between depletion events and BrO episodes.

Conclusions

This atmospheric Hg project in Kuujjuarapik is in its first stage of development and is part of one of the first studies to conduct atmospheric chemistry measurements in northern Quebec. Important findings were made within this first year, 1999-2000. Background Hg in air in Kuujjuarapik is greater than at other remote locations in Canada; however, Hg in precipitation seems to be low, with a south-to-north decreasing concentration gradient.Hg depletion events are extended over the whole Arctic area in the springtime. Fresh Hg deposition is volatilized further in the days following the event and Hg depletion seemed to be connected with BrO episodes.

Expected Project Completion Date

This project is ongoing.

Acknowledgments

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Figure 2. Time series TGM flux, TGM, ozone and net solar radiation measured in Kuujjuarapik, April 14-19, 2000

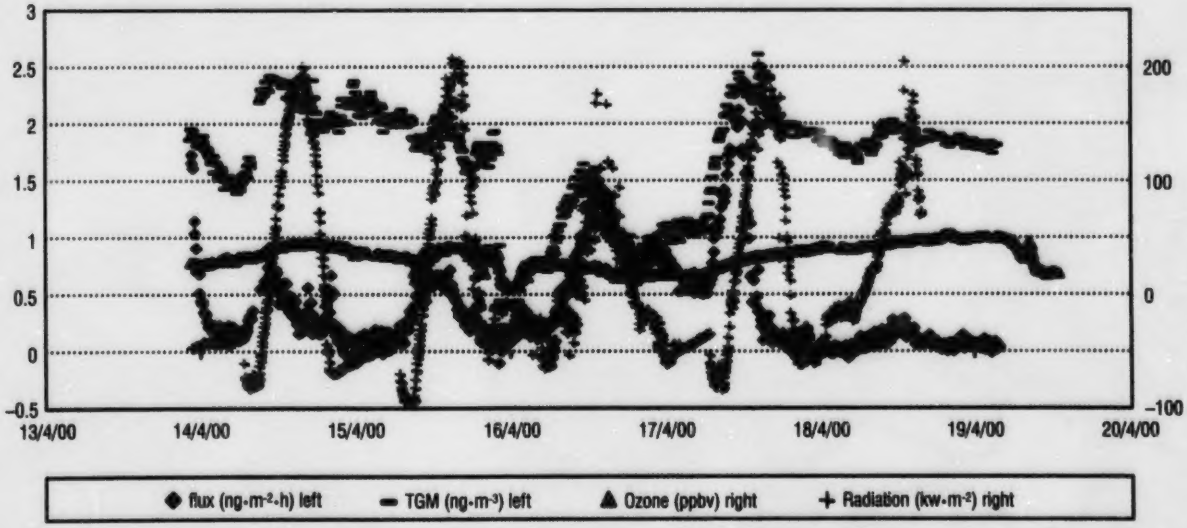
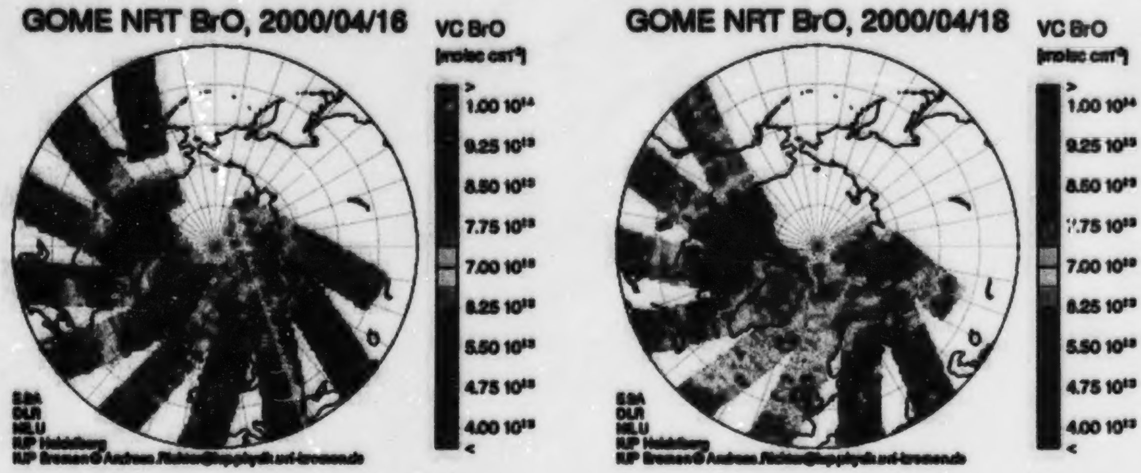


Figure 3. BrO concentration measured by the Global Ozone Monitoring Experiment during a TGM depletion event (April 16, 2000) and a non-depletion event (April 18, 2000) in Kuujjuarapik*


GOME NRT BrO, 2000/04/16
VC BrO
[molec cm-2]
>
1.00 10¹⁴
9.25 10¹³
8.50 10¹³
7.75 10¹³
7.00 10¹³
6.25 10¹³
5.50 10¹³
4.75 10¹³
4.00 10¹³
<
ESA
DLR
NILU
IUP Heidelberg
IUP Bremen
GOME NRT BrO, 2000/04/18
VC BrO
[molec cm-2]
>
1.00 10¹⁴
9.25 10¹³
8.50 10¹³
7.75 10¹³
7.00 10¹³
6.25 10¹³
5.50 10¹³
4.75 10¹³
4.00 10¹³
<
ESA
DLR
NILU
IUP Heidelberg
IUP Bremen

* Source: Global Monitoring Experiment 2000

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Mercury Measurements in Alert

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Abstract

Atmospheric mercury measurements have been made at Alert, Nunavut, since 1995. Baseline data are being collected in order to study spatial and temporal trends of mercury in the Canadian Arctic. This study has discovered that during the springtime, mercury is transformed from the stable species (total gaseous mercury) to a much shorter-lived, more reactive form that can have a significant impact on the Arctic environment. In 1999, total gaseous mercury data were collected and found to show seasonal patterns similar to previous years. Total particulate mercury collected for a 1-year period showed low levels during most of the year and high levels during the polar sunrise mercury depletion events. As well, snow samples collected during the winter and spring of 1998 and 1999 showed increased levels in mercury depositional fluxes during the mercury depletion events, and low fluxes during other times. The implication of these combined findings is that there is a link between the gaseous elemental mercury, oxidized particulate mercury and mercury in precipitation, and ultimately mercury in the aquatic ecosystem. This could help to explain how the remote Arctic environment is being contaminated by this persistent toxic contaminant.

Objectives

1. To collect baseline ambient air concentration measurements of total gaseous mercury (TGM) at Alert, Nunavut for the purpose of establishing:
   - ambient air concentrations in the Canadian Arctic and their linkage to elevated levels of mercury (Hg) in the Arctic food chain
   - temporal and long-term trends in the high Arctic
   - emission sources through long-range transport studies
   - a scientific information basis for use in the development of Canadian objectives for international control strategies
2. To study the chemical and physical aspects of atmospheric Hg vapour transformations after polar sunrise and the resulting link to enhanced concentrations in the Arctic environment.

Introduction

Hg is a toxic metal and has been identified as a priority pollutant by various national and international organisations. Levels of Hg in many Arctic wildlife are elevated (Lockhart 1999), creating a concern among northern communities. In 1995, continuous TGM measurements began in the Canadian Arctic at Alert, Nunavut. Since this time, significant advances in our knowledge of atmospheric Hg in this environment have been made. It was discovered that during the springtime Hg is transformed from the relatively long-lived stable species (TGM) to a much shorter-lived, more reactive form (Schroeder et al. 1998). It is thought that this conversion after the polar sunrise may provide a pathway for the introduction of the more reactive Hg species to the Arctic environment and may be impacting large areas of the Northern Hemisphere.

This program provides long-term data on the spatial and temporal trends of Hg in the Arctic environment. These results can be used for developing and implementing Canadian international strategies, and negotiating positions to achieve appropriate controls for priority pollutants of concern, such as Hg. Results from this program have created international interest and have resulted in similar measurements being made in Alaska, Norway and forthcoming in Russia. This data (Canadian and other) will provide important information on the transport, transformation and deposition processes of this priority pollutant throughout the polar regions.

Activities in 1999-2000

Ground-based atmospheric TGM measurements continued at Alert. The Hg measurement program at Alert is a participant in the Canadian Atmospheric Mercury Measurements Network (CAMNet). Over this past year, this network has implemented the Research Data Management and Quality Control System (RDMQ) for the quality control of the data. The protocols have been applied to the 1997-1999 Alert data sets.

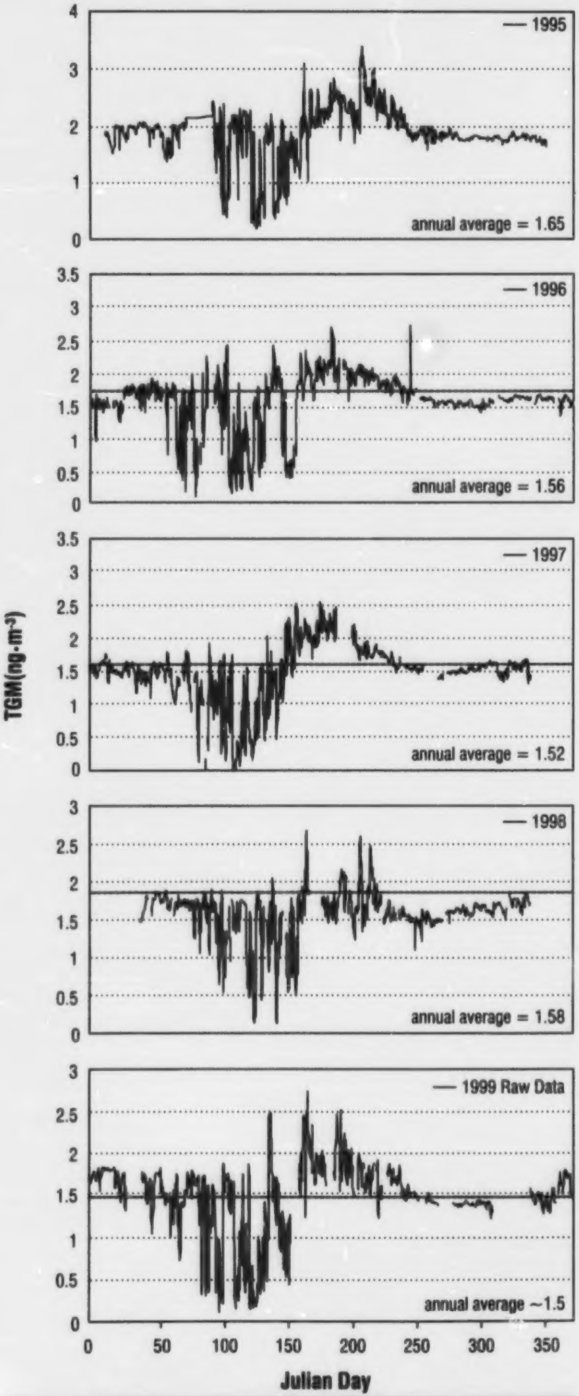
For the first time, an annual study on total particulate mercury (TPM) has been made that provides a further understanding of this species in the Arctic environment

Samples of freshly fallen snow were collected at Alert over a 3-month period during the winter and spring of 1998 and 1999. These samples were collected on a Teflon covered table and analysed using ultra-clean methods for the depositional flux of total Hg.

Results

Ground level measurements were continued for TGM in ambient air at Alert. TGM concentrations for 1995-1999 are shown in Figure 1. The annual pattern for 1999 is similar to the past 4 years. This pattern shows level autumn and winter concentrations, variable springtime concentrations and elevated summer concentrations. Table 1 shows the annual and seasonal statistics for

Figure 1. Annual TGM (ng·m^{-3}) time series (6-hour averaged concentrations), Alert, Nunavut, 1995-1999



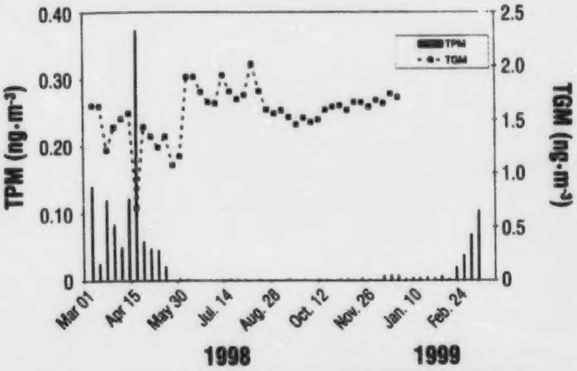
1995-1998 (1999 data are still in quality control completion stages).

An annual time series for TPM was collected and analyzed from March 1998 to the end of February 1999. These results are shown in Figure 2.

Table 1. Annual and seasonal statistics for TGM (ng·m^{-3}), Alert, Nunavut, 1995-1998

	Mean (ng·m^{-3})	Standard Deviation	Range (ng·m^{-3})
Annual 1995	1.65	0.01	0.21- 3.04
Annual 1996	1.56	0.01	0.05 - 4.09
Annual 1997	1.52	0.01	0 - 2.76
Annual 1998	1.58	0.004	0.09 - 3.04
Winter 1995	1.63	0.15	1.10 - 1.94
Winter 1996	1.48	0.41	0.05 - 2.36
Winter 1997	1.48	0.18	0.13 - 2.02
Winter 1998	1.62	0.24	0.43 - 2.04
Spring 1995	1.41	0.59	0.12 - 2.92
Spring 1996	1.40	0.65	0.08 - 2.84
Spring 1997	1.21	0.63	0.00 - 2.58
Spring 1998	1.37	0.50	0.09 - 3.04
Summer 1995	1.89	0.31	1.31 - 3.04
Summer 1996	1.79	0.23	1.45 - 4.09
Summer 1997	1.53	0.08	1.25 - 1.70
Summer 1998	1.67	0.25	0.91 - 2.81
Autumn 1995	1.54	0.04	1.39 - 1.68
Autumn 1996	1.55	0.06	1.31 - 1.97
Autumn 1997	1.53	0.08	1.25 - 1.70
Autumn 1998	1.64	0.09	1.30 - 1.88

Figure 2. Annual time series of TPM and TGM (weekly means), Alert, Nunavut, March 1998-March 1999.



Results from 2 years of depositional fluxes of Hg in the snow phase are shown in Figures 3 and 4. The depositional flux is calculated from the measured Hg concentration, the snow sample area and the mass of the sample.

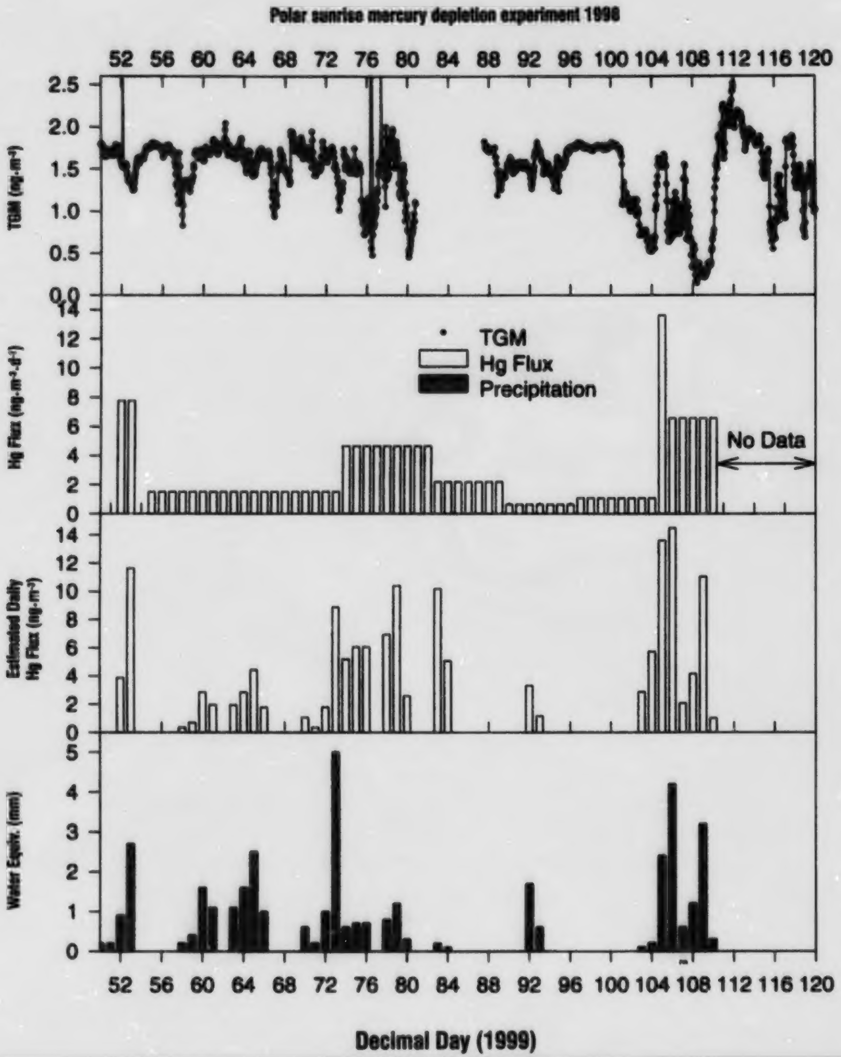
Discussion and Conclusions

Continuous TGM measurements from the past 5 years reveal four distinct seasonal patterns. Fall period TGM concentration levels are very steady (small standard deviation) and consistent from year to year. These are around what is expected to be northern hemispheric background levels (~1.5 ng·m^{-3}). Springtime concentrations are on average lower and highly variable (higher standard deviation and range of the concentrations). This variability is a result of the Hg conversion episodes that result from the polar sunrise sunlight-induced photochemical reactions (Schroeder et al. 1998). Interestingly, summer TGM concentrations are higher than the annual averages and are thought to be a release of Hg that has accumulated in the snow pack during summer melt.

Long-term trends of Hg in the Arctic atmosphere must be addressed, but several more years of data are required for an accurate view. Annual data from Alert are being used to investigate large-scale transport pathways of Hg emitted from source regions (Cheng and Schroeder 2000). This has been applied to the 1992-1993 data as well as the 1995 Alert data. Further investigations using annual data from other years are planned.

The analysis of total particulate Hg was completed using a new technique developed in our laboratory (Lu et al. 1998). The new device is both a particulate trap and pyrolyzer for airborne particulate Hg species. Figure 2 shows the annual time series of TPM and TGM at Alert for samples collected between March 1998 and February 1999. TPM concentrations are elevated in the spring after polar sunrise (March to end of May 1998) with an average of 0.083 ng·m^{-3} (range: 0.021 to 0.37 ng·m^{-3}). The TPM values between June 1998 and February 1999 ranged from 0 to 0.0074 ng·m^{-3}. These values agree with those observed at Ny-Ålesund, Svalbard (Munthe et al. 1997). The elevated TPM values are anti-correlated with TGM and surface ozone concentrations (Schroeder et al. 1998). These results reveal several things. Each spring after polar sunrise, the relatively long-lived TGM comprised mainly of elemental Hg is transformed into much shorter-lived particulate and/or reactive gaseous forms. Since the transformation is strongly correlated with ozone depletion in the atmospheric-marine boundary layer, we believe that Hg^0 is oxidized by reactive species, such as BrO, Br, Cl, BrCl, that are produced in sunlight-induced

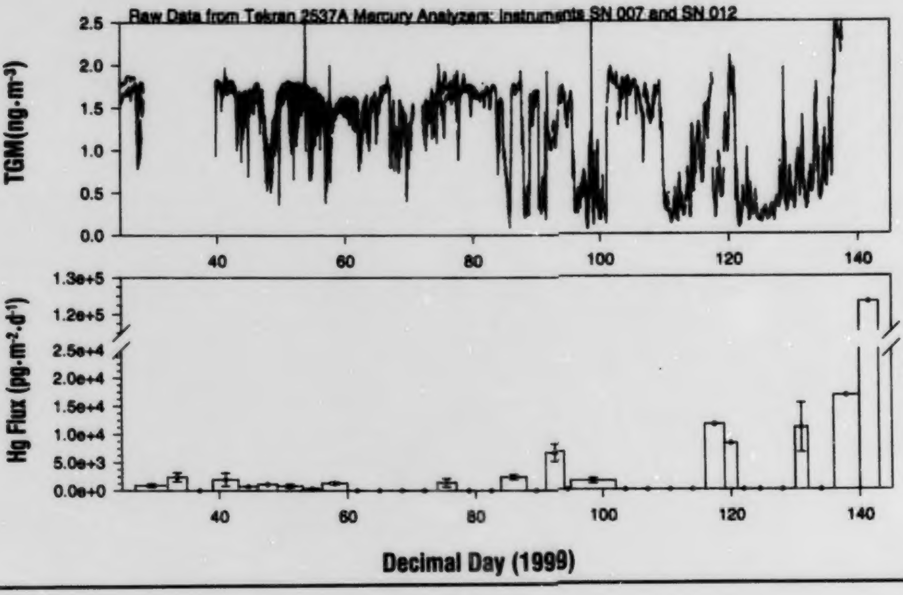
Figure 3. Depositional fluxes of Hg in the snow phase, 1999



photochemical reactions. As a result of the transformation, the depositional flux rate of Hg is enhanced. And finally, during the short northern spring melt, accumulated Hg in the snow pack results in inputs to the biosphere that are many times higher than the mean winter deposition rate.

Experiments to measure the depositional fluxes of Hg in the snow phase have been performed since 1998. Results from 1998 and 1999 are shown in Figures 3 and 4, respectively. During the 1998 period, the flux results show only a qualitative relationship with the TGM depletions. The low sampling frequency (weekly), limited pre-sunrise snow data, and the low number of snow events during the heavy depletion period make the comparison with the TGM data cursory at best. In general, the estimated daily fluxes before the heavy TGM depletion time (day 72) are typically below 4000 pg·m^{-2}·day^{-1} while after day 72 they increase to between 5000 and 14 000 pg·m^{-2}·day^{-1} For the 1999 period, the sampling frequency was increased to every 3 or 4 days while the pre-sunrise interval was sampled much longer to get a better idea of background deposition in the dark winter months. However, like the 1998 season, the number of snow events during the heavy TGM depletion period was infrequent. Even so, a steady increase in the depositional Hg flux is seen after about day 80, corresponding to the start of the heavy TGM depletion events. Deposition rates increase from < 2500 pg·m^{-2}·day^{-1} during the dark, pre-sunrise period to between 6500 and 17 000 pg·m^{-2}·day^{-1} during the heavy TGM depletion period. An enormous flux rate of 120 000 pg·m^{-2}·day^{-1} was observed around day 140 in 1999, corresponding to a huge snow Hg concentration

Figure 4. Depositional fluxes of Hg in the snow phase, 1999



TGM and Hg depositional flux results for Alert, NWT: PSMDE99

(almost 2.6 ng·g^{-1} versus the dark period mean snow Hg concentration of 4.9 pg·g^{-1}) but only a small amount of snow deposition. The incident may be a washout effect, often seen during rain events, where the majority of the scavenged Hg comes out in the first fraction of the rain (Bloom and Watras 1989, Glass et al. 1991).

The implication of these combined findings is that there is a link between the gaseous elemental Hg phase, oxidized particulate Hg and Hg in precipitation, and ultimately Hg in the aquatic ecosystem. This could help to explain how the remote Arctic environment is being contaminated by this persistent toxic contaminant.

Expected Project Completion Date

Ongoing

Acknowledgments

The project team would like to thank the Global Atmospheric Watch program at Alert for supplying facilities, assistance and personnel. We would like to thank Erin Tait, Chris Shelswell and Jacinta McNairn for collecting the 1999 TGM data and snow samples. Also, the facilities and assistance from the Department of National Defense and the continuous support of the personnel at CFS Alert have been invaluable and is truly appreciated.

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Quantifying and Reducing Uncertainty in Model Calculations of Global Pollutant Fate

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Abstract

As part of the overall objective to quantify the global distribution behaviour of persistent organic pollutants using model calculations, semi-empirical estimates of the two processes deemed most significant for the loss of polychlorinated biphenyls (PCBs) from the global environment were made. The estimate of the loss of PCBs and hexachlorocyclohexanes by reaction with hydroxyl radicals takes into account congener-specific degradation rates, the effect of partitioning onto aerosols, and the spatial and temporal variability of temperature, hydroxyl radicals and contaminant concentrations. The lifetimes due to reaction with hydroxyl radicals in the global atmosphere likely range from about 1 month for the light PCBs to in excess of 1 year for the heavy congeners. This implies a global annual loss of PCBs during the past 30 years in the range of hundreds to thousands of tonnes, most of it due to reactions of the lighter congeners. The global loss of PCBs by settling with organic particles to the deep sea is likely on the same order of magnitude as the atmospheric degradation, but affects the more highly chlorinated congeners more than the lighter ones.

Objectives

The main objective of the project is to describe quantitatively with model calculations the global distribution behaviour of persistent organic contaminants and to establish credibility in the results of these simulations. Specifically, the following tasks were foreseen for the project year 1999-2000:

1. Bring to completion and seek to publish a comprehensive study of the global fate of PCBs using the zonally averaged global distribution model, with a particular focus on changes in the relative homologue composition with latitude and between different compartments;
2. Conduct a semi-empirical estimation of global atmospheric degradation, atmospheric deposition to the world oceans, and deep sea transfer for PCBs and HCHs (hexachlorocyclohexanes) using measured concentrations in air and ocean water and process descriptions implemented in the global distribution model; and

3. Conduct a comprehensive uncertainty analysis of the global model calculations for α-HCH and PCBs to increase credibility in the descriptive and predictive capabilities of the model.

Introduction

The ultimate goal of this project is the quantitative description of the global fate of persistent organic pollutants (POPs). Of particular importance for the Northern Contaminants Program (NCP) is the question of future time trends in the Arctic. How long will it take for contamination levels in the Arctic food chain to decrease to acceptable levels? Since POP concentrations in the Arctic reflect background levels away from sources, the rate of decrease of a pollutant in the Arctic is tied to the rate of clearance of that pollutant in the global ecosystem as a whole. The clearance rate of the global environment can be expressed with the help of an average global lifetime τ_{tot} defined as M_{tot}/N_{tot} where M_{tot} is the total amount of a pollutant dispersed in the global environment and N_{tot} is the sum of all loss processes. A loss process in this context is a process that leads to a permanent removal of the chemical from the sphere that is accessible to organisms. For persistent organic pollutants, such removal may occur by:

- degradation in various environmental compartments
- transfer to the deep sea with settling particulate matter and down-welling water
- burial and irreversible sorption in freshwater and marine shelf sediments
- leaching into the ground and irreversible sorption in the terrestrial environment
- loss to the stratosphere
- burial in glaciers, icecaps and peat bogs.

For PCBs, only two of these mechanisms are suspected to contribute significantly to the rate of loss from the global environment, namely the reaction of gaseous PCBs with the hydroxyl (OH) radical in the atmosphere and the export to the deep sea with settling particulate organic matter. Any model that aims to describe and predict the trends of PCB concentrations in the global environment over the time scale of decades needs to accurately account for these two processes. As part of our overall modelling study we thus studied in detail how to describe these processes on a zonally averaged basis.

Activities in 1999-2000

This synopsis report will focus on the results for Objective 2 stated above. The publication of the model results of the global fate of PCBs (Wania 1999, Wania et al. 1999a) was held, pending the release of the first publicly distributed version of the global distribution model (Wania and Mackay 2000) and the completion of the semi-empirical process estimations (see below). The results of these estimations allow a check of the reasonableness of the process descriptions and results of the global distribution model for PCBs. Work on Objective 3 is still ongoing and will be described in detail in next year's synopsis report. Four processes of significant impact on the global fate of PCBs were investigated: uptake in forests, air-soil exchange, atmospheric degradation and deep-sea transfer. The results of the assessment of the importance of forests in the global fate of POPs are summarized in a recently submitted manuscript (Wania and McLachlan 2000). Another manuscript on modelling the exchange of POPs between air and soil is nearing completion (McLachlan and Wania 2000). Instead of recounting what is in these two manuscripts, we describe here the results of the semi-empirical estimates for atmospheric degradation and deep-sea transfer.

Results and Discussion

Estimating global loss of PCBs and HCHs by atmospheric degradation

The description of the loss of PCBs and HCHs by degradation in the atmosphere appears at first to be simple and unproblematic. It is fairly well established that the reaction with OH radicals is the only significant reaction of these chemicals in the atmosphere (Atkinson 1996), and as a result of the work by the group of Atkinson, reaction rates of individual PCB isomers with OH radicals are well established (Atkinson and Aschman 1985, Kwok et al. 1995). These rates have further been confirmed by independent measurements by Anderson and Hites (1996a, 1996b), who also derived information on the temperature dependence of these reactions. Brubaker and Hites (1998) reported such information for α- and γ-HCH. As a result of the work by these two groups, we thus can calculate the degradation rate k_A in s^{-1} at a particular temperature T (in K) and OH radical concentration [OH] (in molecules·cm^{-3}) from:

$$k_A = k_{Aref} \cdot [OH] \cdot e^{\frac{E_a}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)}$$

Table 1. Chemical properties of five PCB congeners used in the estimation of atmospheric lifetimes due to reaction with OH radicals

	PCB-28	PCB-52	PCB-101	PCB-153	PCB-180	α-HCH	γ-HCH
k_{WAref} in $cm^3 \cdot molecules^{-1} \cdot s^{-1}$*	$1.04 \cdot 10^{-12}$	$5.90 \cdot 10^{-13}$	$3.00 \cdot 10^{-13}$	$1.60 \cdot 10^{-13}$	$1.00 \cdot 10^{-13}$	$1.4 \cdot 10^{-13}$	$1.9 \cdot 10^{-13}$
log K_{OAref} $^{(a)}$	7.6	7.72	8.38	9.09	9.72	7.39	7.76
ΔH_{OA} in $kJ \cdot mol^{-1}$	–82.8	–88.7	–94.2	–99.4	–103.3	–78.1	–72.5

* at reference temperature 25°C.

where k_{Aref} is the reaction rate at the reference temperature T_{ref} in $cm^3 \cdot molecule^{-1} \cdot s^{-1}$, R is the gas constant and E_A is the activation energy of the reaction in $J \cdot mol^{-1}$. Table 1 lists k_{Aref} values, which in the case of PCBs were derived from the relationship reported in Atkinson (1991) and in the case of HCHs are the reported experimental values (Brubaker and Hites 1998).

The validity of these laboratory-derived reaction rates in the real atmosphere, however, has been questioned repeatedly. Axelman (1997) concluded from global budget estimates that these reaction rates may be too high, possibly by more than an order of magnitude. Scheringer (1997) stated that the observed long-range transport behaviour of semi-volatile organochlorines is not reproduced by models relying on gas-phase degradation rates alone. He suggested that adsorption to aerosols might preclude some POPs from being degraded.

This issue is of particular relevance in the context of the emerging need to classify chemicals as being subject to long-range transport. The degradation rate in the atmosphere is the single most important parameter controlling the atmospheric travel distance of organic chemicals (e.g. Beyer et al. 2000), and the question of whether laboratory derived OH reaction rates have real world applicability is thus of utmost importance. To explore this issue, we have made several complementary, increasingly complex estimates of the loss of PCBs and HCHs from the atmosphere by reaction with OH radicals. We again resort to the use of an average lifetime, this time the mean atmospheric lifetime τ_A of a pollutant, which is the amount of chemical in the entire atmosphere M_A divided by the total loss by atmospheric degradation N_A.

$$\tau_A = \frac{M_A}{N_A}$$

Estimate 1: Using averaged OH radical concentrations and temperatures

If we assume that the entire amount of chemical present in the atmosphere participates in the degradation reaction with OH radicals, τ_A is simply the reciprocal of the rate constant k_A.

$$\tau_A = \frac{M_A}{M_A \cdot k_A} = \frac{1}{k_A}$$

We can estimate k_A by applying a spatially and temporally averaged [OH] and T to the measured congener specific laboratory derived reaction rates k_{Aref}. Using this approach, Atkinson's group estimated atmospheric lifetimes τ_A for PCBs of variable degree of chlorination (Kwok et al. 1995). Applying an OH radical concentration of $8.0 \cdot 10^5$ $molecules \cdot cm^{-3}$ and room temperature rate constants, they obtained the values listed in Table 2, suggesting lifetimes from a few days for light PCBs to several months for heavily chlorinated PCB congeners. These values do not take into account that the reaction with OH radicals is temperature dependent (Anderson and Hites 1996b). The global atmosphere is on average a lot colder than room temperature. Using temperature values implemented in the global distribution model (Wania et al. 1999a), we calculated a spatially (density-weighted) and annually averaged atmospheric temperature value of –18°C.

Table 2. Atmospheric lifetimes (τ_A) due to reaction of PCBs of various degrees of chlorination with OH radicals* assuming room temperature, no partitioning onto aerosols and a OH radical concentrations of $8 \cdot 10^5$ cm^{-3}

No. of Chlorine	1	2	3	4	5	6
τ_A in days	2.7 - 5.1	3.4 - 7.2	6.9 -15	8.5 - 40	16 - 48	29 - 90

* Estimated by Kwok et al. 1995

The globally and annually averaged OH radical concentration in the earth's atmosphere is by no means an established figure, but the numbers being reported tend to be in the range $4 \cdot 10^5$ to $10 \cdot 10^5$ radicals·cm^{-3}. Altshuller (1989) reported an annually and diurnally averaged northern hemispheric average of $4 \cdot 10^5$ cm^{-3}. Other global tropospheric means of $8 \cdot 10^5$ and $10.3 \cdot 10^5$ cm^{-3} are reported by Spivakovsky et al. (1990) and Hein et al. (1997). From measured trichloroethane concentrations, Prinn et al. (1995) derived a temperature and atmospheric density weighted average of $(9.7 \pm 0.6) \cdot 10^5$ cm^{-3}, which was adopted by Anderson and Hites (1996b) in their estimate of PCB loss from the atmosphere. From the two-dimensional, seasonally changing model results for [OH] by Rodriguez et al. (1992), which are implemented in the global distribution model (Wania et al. 1999a), we derived a spatially (density weighted) and annually averaged value of $7.25 \cdot 10^5$ cm^{-3}. Being in the middle of the range reported, we used this value for our estimates of τ_A. This also facilitates a direct comparison with the τ_A estimates derived from the global distribution model (see below). However, it should be kept in mind that there is an uncertainty in this number, which directly (linearly) affects the estimated degradation rates.

Using the rate constants k_{Aref} from Table 1 and assuming an activation energy of the reaction of 10, 11.2 and 14.2 kJ·mol^{-1} for the PCBs, α-HCH and γ-HCH, respectively, and a [OH] of $7.25 \cdot 10^5$ cm^{-3}, we estimated the atmospheric lifetimes for five selected PCB congeners and the two HCH isomers at three temperatures (Table 3, Estimate 1). At 25°C the lifetimes are similar, if somewhat longer than those estimated by Kwok et al. (1996). At lower temperatures, however, the lifetimes are considerably longer. Nevertheless, even at the average atmospheric temperature of −18°C the estimated lifetimes are shorter than one year for all congeners.

Estimate 2: Taking into account partitioning between vapour and particle phase

The most serious limitation of the above estimates is that they fail to account for the tendency of many PCBs to partition between the vapour phase and the atmospheric particle phase (Scheringer 1997). Whereas the degradation rates measured by the groups of Atkinson and Hites are for the gas phase, the reactions of organic contaminants sorbed onto aerosol particles are poorly understood, at least from a quantitative point of view. Only recently, experimental studies have been conducted, but none involving PCBs or HCHs (Palm et al. 1997, Palm et al. 1999).

If we assume that a chemical that is sorbed to aerosols does not participate in the reaction with OH radicals, we obtain:

$$\tau_A = \frac{M_A}{N_A} = \frac{V_A \cdot C_{Atot}}{V_A \cdot C_{Agas} \cdot k_A} = \frac{C_{Agas} + C_{Apart}}{C_{Agas} \cdot k_A}$$

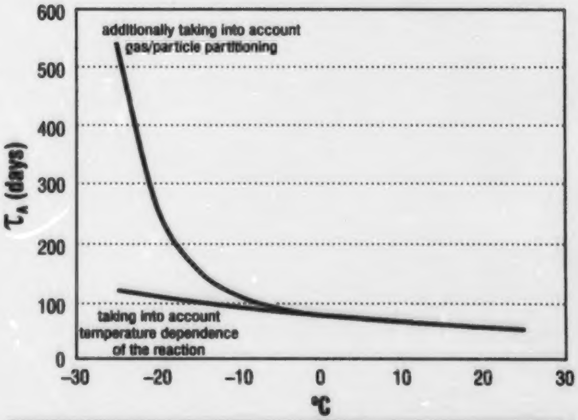
where C_{Agas} and C_{Apart} are the concentration of the chemical in the gas and particle-sorbed phase, respectively. If we further assume that partitioning onto aerosols is well-described with the empirical relationship derived by Finizio et al. (1997), we obtain:

$$\tau_A = \frac{1 + 3.5 \cdot K_{OA} \cdot V_Q}{k_A}$$

where v_Q is the volume fraction of aerosols in the atmosphere, K_{OA} is the octanol-air partition coefficient and 3.5 is an empirical parameter relating the aerosol-air partition coefficient with K_{OA} (Finizio et al. 1997). Assuming that a typical background aerosol concentration is $2 \cdot 10^{-12}$ m^3·m^{-3} (Bidleman 1988), this implies that partitioning to aerosols has an impact on the atmospheric lifetime of compounds with log K_{OA} in excess of 10 ($3.5 \cdot K_{OA} \cdot v_Q = 0.07$ for log $K_{OA} = 10$ and 0.7 for log $K_{OA} = 11$). At 25°C, only a few, highly chlorinated PCB isomers have a K_{OA} high enough to partition significantly onto aerosols. However, K_{OA} is highly temperature dependent and the atmosphere is a lot colder than 25°C at higher latitudes and altitudes. The effect of temperature on τ_A is illustrated in Figure 1. Obviously, gas-particle partitioning greatly attenuates the effect of temperature on OH radical reaction.

If we recalculate τ_A for the five selected PCB congeners and the two HCH isomers, taking into account the partitioning onto aerosols and using congener-specific

Figure 1. Estimated atmospheric lifetime (τ_A) of PCB-101 in the temperature range −25°C to 25°C, assuming an [OH] of $7.25 \cdot 10^5$ cm^{-3}



temperature-dependent K_{OA} values (Table 1), we yield the values in Table 3 (Estimate 2). At 25°C, the lifetimes are essentially identical to those from Estimate 1. Whereas the effect of introducing gas/particle partitioning on the atmospheric lifetime of the lighter PCBs and the HCHs at 0°C is still insignificant, the lifetime of PCB-180 is now more than doubled. At −18°C the lifetime of all PCB congeners is strongly affected by gas/particle partitioning, and the heavier congeners become completely persistent with lifetimes in excess of one year.

Atmospheric temperatures in the upper atmosphere are generally lower than −25°C. Virtually all PCBs will be sorbed to particles at these temperatures and will thus not participate in the gas phase reactions with OH radicals. This has generally been overlooked in earlier estimates of global or hemispheric lifetimes of PCBs,

Table 3. Atmospheric lifetimes due to reaction with OH radicals τ_A for five PCB congeners, estimated using five different methods.

	PCB-28	PCB-52	PCB-101	PCB-153	PCB-180	Sum of 5	α-HCH	γ-HCH
No. of chlorine	3	4	5	6	7		6	6
Estimate 1: Spatially and temporally averaged air temperature, T, and OH radical concentrations [OH] of 7.25×10^5 cm^{-3}; no partitioning onto aerosols								
25°C	15 days	27 days	53 days	100 days	160 days		114 days	84 days
0°C	22 days	39 days	77 days	144 days	231 days		172 days	142 days
−18°C*	**30 days**	**53 days**	**105 days**	**197 days**	**315 days**		**244 days**	**221 days**
Estimate 2: Like Estimate 1, but temperature dependent partitioning onto aerosols								
25°C	15 days	27 days	53 days	101 days	166 days		114 days	84 days
0°C	22 days	40 days	81 days	193 days	1.7 years		172 days	143 days
−18°C*	**33 days**	**62 days**	**212 days**	**4.5 years**	**>10 years**		**244 days**	**233 days**
Estimate 3A: Spatially variable [OH] and T, temperature dependent partitioning onto aerosols, assuming uniform fugacity								
Air layer 1	15.6 days	27.5 days	56.1 days	131.8 days	1.2 years		115 days	85 days
Air layer 2	17.2 days	31.3 days	85.8 days	1.4 years	>10 years		125 days	93 days
Air layer 3	295 days	4 years	>10 years	>10 years	>10 years		2.5 years	1.9 years
Air layer 4	>10 years	>10 years	>10 years	>10 years	>10 years		>10 years	>10 years
Total amosphere	**7.1 years**	**>10 years**	**>10 years**	**>10 years**	**>10 years**		**>10 years**	**8.3 years**
Estimate 3B: Spatially variable [OH] and T, temperature dependent partitioning onto aerosols, assuming uniform total air concentration								
Air layer 1	15.1 days	26.6 days	52.6 days	100 days	167 days		112 days	83 days
Air layer 2	16.2 days	28.6 days	57.1 days	115 days	227 days		120 days	89 days
Air layer 3	43.6 days	84.9 days	317 days	7.5 years	>10 years		304 days	228 days
Air layer 4	2.8 years	11.8 years	>10 years	>10 years	>10 years		9.9 years	7.2 years
Total atmosphere	**57 days**	**107 days**	**267 days**	**1.8 years**	**3.7 years**		**1.1 years**	**0.8 years**
Estimate 4: Using global distribution model with global historic emission estimates								
M_A (1970) in t	43 t	61 t	73 t	41 t	8 t	226 t	3.8 kt	—
N_A (1970) in $t\cdot a^{-1}$	351 t/a	344 t/a	227 t/a	60 t/a	4 t/a	986 t/a	10.5 kt	—
τ_A (1970)	**44 days**	**65 days**	**117 days**	**251 days**	**1.7 years**	**84 days**	**133 days**	—
M_A (1999) in t	2.2 t	3.3 t	4.4 t	2.5 t	0.4 t	13 t	4.3 t	—
N_A (1999) in $t\cdot a^{-1}$	17 t/a	17 t/a	13 t/a	3 t/a	0.2 t/a	50 t/a	12.4 t	—
τ_A (1999)	**49 days**	**70 days**	**122 days**	**260 days**	**1.8 years**	**92 days**	**126 days**	—

* Global atmospheric average

likely leading to a significant overestimation of this global loss process. Based on similar rate constants for PCBs and propane for the gas-phase reaction with OH radicals, Anderson and Hites (1996b) suggested that the vertical gradient of PCB concentrations in the atmosphere can be approximated by that of propane. However, this is unlikely to be the case since the volatile propane will partition onto particles to a much smaller extent than the semi-volatile PCBs.

Estimate 3: Taking into account the temporal and spatial variability of temperature and OH radical concentration

Obviously, both atmospheric temperatures and OH radical concentrations are not homogeneous in space and time, but rather undergo diurnal and seasonal fluctuations and vary with altitude and latitude. The use of spatially and temporally averaged values will result in errors in the estimated atmospheric lifetimes. Differences in atmospheric [OH] and T are most pronounced on a vertical and meridional scale, whereas they tend to be relatively homogeneous on a zonal scale. We thus used the 40 atmospheric boxes of the zonally averaged global distribution model (Wania et al. 1999a) to estimate the effect of that variability on τ_A. In this model, the global atmosphere is divided into four vertical layers and 10 latitudinal zones. For each of these 40 boxes, monthly values of [OH] and T are defined based on information in Rodriguez et al. (1992) and Fleming et al. (1990).

The tropospheric half-life in a compartmentalised atmosphere is calculated as the sum of the amounts in each compartment i divided by the sum of the loss in each compartment i:

$$\tau_A = \frac{\sum_i M_{Ai}}{\sum_i N_{Ai}} = \frac{\sum_i V_i \cdot C_{Atot_i}}{\sum_i V_i \cdot C_{Agas_i} \cdot k_{Ai}}$$

Each compartment i with volume V_i may have different $[OH]_i$, T_i and contaminant concentrations C_{Atoti} and C_{Agasi}. In order to calculate τ_A with the above equation, it would be necessary to specify contaminant concentrations C_{Atoti} and C_{Agasi} in each of the 40 compartments, preferably as a function of time. Our knowledge of PCB and HCH concentrations in the global atmosphere, especially in higher altitudes and in the southern hemisphere, is too limited to do this with great confidence.

By making certain simplifying assumptions, it is possible to calculate lifetime estimates for the whole atmosphere, or parts of it, without specifying the spatial and temporal distribution of contaminants in the atmosphere. We consider two such simplifications:

We first assume that the whole atmosphere is in chemical equilibrium, implying a constant fugacity f_{Ai} throughout the 40 atmospheric compartments. Then:

$$\tau_A = \frac{\sum_i V_i \cdot C_{Atot_i}}{\sum_i V_i \cdot C_{Agas_i} \cdot k_{Ai}} = \frac{\sum_i V_i \cdot Z_{BA_i} \cdot f_{A_i}}{\sum_i V_i \cdot Z_{A_i} \cdot f_{A_i} \cdot k_{Ai}} = \frac{f_A \cdot \sum_i V_i \cdot Z_{BA_i}}{f_A \cdot \sum_i V_i \cdot Z_{A_i} \cdot k_{A_i}} = \frac{\sum_i V_i \cdot Z_{BA_i}}{\sum_i V_i \cdot Z_{A_i} \cdot K_{A_i}}$$

where Z_{Ai} and Z_{BAi} are Z-values for the vapour and bulk air phase, respectively. This corresponds to the calculation of a characteristic time V·Z / D value (Mackay 1991). Using again the relationship proposed by Finizio et al. (1997), we obtain:

$$\tau_A = \frac{\sum_i V_i \cdot (1 + 3.5 \cdot K_{OA}(T_i) \cdot V_Q)}{\sum_i V_i \cdot k_{Ai} / (R \cdot T_i)}$$

Using this equation we calculated τ_A for the four atmospheric layers and for the atmosphere as a whole. Twelve monthly values were averaged and are reported in Table 3 (Estimate 3A). For lack of better data, v_Q was assumed to be $2 \cdot 10^{-12}$ at the Earth's surface and the aerosol mixing ratio was assumed to be constant with altitude. The calculations yield extremely long total atmospheric lifetimes, in excess of 5 years for all five PCB congeners and both HCH isomers, suggesting that the assumption of equal fugacity is unreasonable. In reality, air that is rising and thus experiencing a large decrease in temperature will see a significant drop in the fugacity of semi-volatile chemicals as they partition increasingly onto aerosols at the lower temperatures (whose Z-value increases significantly at lower temperature).

Alternatively, we may assume that the total concentration of PCBs or HCHs C_{Atoti} (or more precisely the mixing ratio) is constant throughout the atmosphere. We can then derive:

$$\tau_A = \frac{\sum_i V_i}{\sum_i V_i \cdot k_{Ai} / (1 + 3.5 \cdot K_{OA}(T_i) \cdot V_Q)}$$

Using this approach, we again calculated the average of 12 monthly values for each layer and the atmosphere as a whole and present these τ_A values in Table 3 (Estimate 3B). The atmospheric lifetimes now range from approximately 2 months for PCB-28 to almost 4 years for PCB-180. The HCHs have lifetimes in the range of 1 year. Degradation in higher atmospheric layers is negligible because virtually all of the contaminants are in the particle phase at the low temperatures prevalent at these heights.

Estimate 4: Taking into account the spatial variability of the PCB mixing ratio.

Although we have difficulties specifying PCB and HCH concentrations (or alternatively fugacities) in each of the 40 atmospheric compartments, we know quite well that contaminant concentrations are not the same all over the globe. Based on a literature review, Axelman (1997), for example, suggests that PCB concentrations tend to decrease from low to high latitudes within the northern hemisphere. To estimate the influence of the spatial and temporal variability of atmospheric PCB and HCH concentrations on the τ_A estimate, we employed the latest version of the global distribution model (public release version 1.0, Wania and Mackay 2000). Using the PCB emission estimates by Sweetman and Jones (Wania et al. 1999b) and the α-HCH emission estimates by Li (Wania et al. 1999a), we calculated for the five selected PCB congeners and for α-HCH the amounts M_A and the annual degradation loss N_A in the atmosphere for the years 1970 and 1999. τ_A is calculated as the ratio of these two model results. The results are given in Table 3 (Estimate 4). No such calculation was performed for γ-HCH due to the lack of historical emission estimates.

The τ_A values from Estimate 4 are somewhat lower than those from Estimate 3B. This reflects the fact that the PCB and HCH concentrations are higher in regions of high degradation potential than in regions of limited degradation potential. In other words, air concentrations are higher at low latitudes (especially in the temperate and subtropical zones of the Northern hemisphere, which receive the bulk of the PCB emissions) and altitudes, where lifetimes are shorter than at high altitudes and latitudes, where it is darker and colder, and degradation thus is slower. An interesting result is that the τ_A values of the PCBs are somewhat longer in 1999 than in 1970. This suggests that during this time period the relative distribution of PCBs in the global atmosphere has shifted towards regions of lower degradation potential, most likely towards higher latitudes.

This last method also allows an estimate of a lifetime for the sum of the five PCB congeners. This τ_A is 84 to 92 days and thus closer to that of the lighter PCB congeners, confirming that the loss of PCBs in the atmosphere is dominated by the reaction of the less chlorinated congeners.

The absolute loss for the five congeners by reaction with OH radicals is estimated to be approximately 1000 $t{\cdot}a^{-1}$ in 1970 and 50 $t{\cdot}a^{-1}$ in 1999, reflecting a dramatically decreased air concentration in the intervening 30 years. This compares with the estimate of 8271 $t{\cdot}a^{-1}$ for total PCBs by Anderson and Hites (1996b), which is based on global average surface concentration of 200 $pg{\cdot}m^{-3}$ total PCBs.

Settling of PCBs with particles to the deep sea

Estimating the loss of PCBs with particles settling beyond the halocline in the open ocean is far less straightforward than the loss by reaction with OH radicals. It is particularly difficult due to the very small number of reliable quantifications of PCBs in open sea water. To approach this problem, we used three alternative approaches.

Estimate 1: Upscaling of measured PCB fluxes to the deep ocean

There is a very small number of studies reporting direct measurements of the flux of PCBs to the deep ocean. Knap et al. (1986) reported mean daily PCB fluxes of $4.4{\cdot}10^{-9}\,g{\cdot}m^{-2}{\cdot}day^{-1}$ (range: 1.2 to $10.8{\cdot}10^{-9}\,g{\cdot}m^{-2}{\cdot}day^{-1}$) in the deep Sargasso Sea (North Atlantic) between 1978 and 1980. Dachs et al. (1996) measured $15.1{\cdot}10^{-9}\,g{\cdot}m^{-2}{\cdot}day^{-1}$ (range 6.9 to $24.9{\cdot}10^{-9}\,g{\cdot}m^{-2}{\cdot}day^{-1}$) in the Alboran Sea (western Mediterranean) in spring 1992. More recent measurements by Gustafsson et al. (1997) in the North Atlantic and by Dachs et al. (1999) for the Arabian Sea give somewhat lower values but are in the same order of magnitude. Considering the variability of methodology (e.g. depth of sediment traps) and relative location and distance to source regions, these data span over a surprisingly small range. In particular, we would expect the fluxes to be much higher in the earlier studies than those measured in the 1990s. We also may suspect PCB fluxes in the southern hemisphere to be smaller, considering that use of PCBs there has been much more limited than in the Northern hemisphere. As daring as it may be, we estimated based on these studies that 1 to 5 $\cdot 10^{-9}\,g{\cdot}m^{-2}{\cdot}day^{-1}$ may be a realistic figure for the average present day flux of PCBs to the deep ocean. This would translate to global annual PCB losses to the deep sea of approximately 100 to 700 tonnes.

Estimate 2: Estimating PCB concentrations on suspended organic carbon particles.

Most studies measuring PCB fluxes to the deep sea observe a correlation with the flux of particulate organic carbon (POC). It should thus be possible to estimate the flux of PCBs to the ocean by multiplying the average concentration of PCBs in POC in the open sea with the flux of particulate organic carbon to the deep sea. The latter is a relatively well-known parameter because it is of such eminent importance to the global carbon cycle. A recent estimate is 16 gigatonnes of organic carbon per year (Falkowski et al. 1998). The former is notoriously difficult to measure, due to the low concentration of POC in the ocean and the low concentrations of PCBs in the POC. Reported measurements may thus be highly uncertain. Based on studies by Knap et al. (1986), Dachs et al. (1996), Schulz-Bull et al. (1998) and others, we suggest that a typical particulate PCB concentration in the open ocean is $1 \cdot 10^{-7}$ g PCB (= 100 ng) per g of POC. When combined with the carbon flux, this yields estimates of the loss of PCB of 1600 tonnes per year, i.e. almost an order of magnitude higher than those obtained using the method for Estimate 1.

Estimate 3: Estimating the PCB load in the deep ocean

An entirely different approach to estimating the loss of PCBs to the deep sea is to estimate the inventory of PCBs in the deep ocean and to infer what rate of settling would supply this amount of PCBs. As most settling organic matter decomposes before reaching the deep ocean floor, PCBs sorbed to the organic matter will be released to the water column and are not suspected to accumulate in the deep sea sediments (Axelman 1997). However, we are not aware of any studies reporting PCB concentrations in deep ocean sediments that would allow us to confirm this. Taking the measurements by Schulz et al. (1988) in the North Atlantic as an indication of the contamination of the world's entire deep ocean, we may assume an average PCB concentration of 1 $ng \cdot m^{-3}$. This translates into a total inventory of 1500 tonnes of PCBs in the deep ocean. Considering that PCBs have been emitted for many decades and fluxes were likely much higher in the 1960s and 1970s than they are now, this seems too low when compared to the fluxes estimated above. We are left to speculate whether the estimates based on PCB and POC fluxes are too high, the water column concentrations are higher than 1 $pg \cdot L^{-1}$ or whether deep sediment may after all be a repository for PCBs delivered to the deep sea (Froescheis et al. 2000). Axelman (1997) also noted this inconsistency between the import to, and the standing stock of, PCBs in the deep sea.

Estimate 1, which is the most direct method, gives a flux estimate which lies in between the flux estimates derived with the two indirect methods 2 and 3, and may thus be the most reasonable. It should be obvious that such estimates are highly uncertain, indicating nothing more than the correct order of magnitude of that global loss process.

Particle settling in the global distribution model

We compared the results of these three estimation methods with the fluxes of five PCB congeners to the deep sea calculated with the global distribution model. The model derives a contaminant loss rate to the deep sea by calculating a contaminant concentration on particulate matter within the surface ocean and applying a particle settling rate U_{9ocean} in units of $m \cdot h^{-1}$ (Wania and Mackay 1995). It is thus somewhat similar to the method for Estimate 2. Originally, a particle settling rate of $5 \cdot 10^{-8}$ $m \cdot h^{-1}$ in temperate to tropical oceans was used, and somewhat lower rates in the high latitude oceans (Wania and Mackay 1995). Later a uniform particle settling rate of 10^{-7} $m \cdot h^{-1}$ was employed (Wania 1998, Wania et al. 1999a). A study using the global distribution model to estimate the long-range transport behaviour of a whole range of chemical property combinations indicated however that these particle settling rates overestimate transport to the deep sea (Wania 1998). In the case of modelling the global fate of α-HCH, this weakness did not affect model performance to a significant extent because this process is not very important for the relatively water soluble α-HCH (Wania et al. 1999a). As an interim solution, a uniform settling rate of 10^{-8} $m \cdot h^{-1}$ was used in preliminary PCB fate simulations (Wania et al. 1999b). The public release version of the model (Wania and Mackay 2000) uses zonally variable flux rates derived from information in Falkowski et al. (1998). Using the global map of carbon export to the deep sea in that publication, we estimated the average carbon flux in each of the 10 zones of the model, and from these deduced particle settling rates in $m \cdot h^{-1}$ (Table 4).

Using this latest model version, the annual loss of the sum of five PCB congeners (28, 52, 101, 153, 180) to the deep sea was estimated to be 362 tonnes in 1970 and 24 tonnes in 1999, most of which was due to the transport

Table 4. Oceanic particle settling rates*

Northern hemisphere	**Polar**	**Boreal**	**Temperate**	**Subtropic**	**Tropic**
Average organic carbon export ($g\ OC \cdot m^{-2} \cdot a^{-1}$)	75	55	121	44	33
Particle settling rate ($m \cdot h^{-1}$)	$2.25 \cdot 10^{-8}$	$1.65 \cdot 10^{-8}$	$3.60 \cdot 10^{-8}$	$1.32 \cdot 10^{-8}$	$1.00 \cdot 10^{-8}$
Southern hemisphere	**Polar**	**Subpolar**	**Temperate**	**Subtropic**	**Tropic**
Average organic carbon export ($g\ OC \cdot m^{-2} \cdot a^{-1}$)	5	33	55	55	33
Particle settling rate ($m \cdot h^{-1}$)	$0.15 \cdot 10^{-8}$	$1.00 \cdot 10^{-8}$	$1.65 \cdot 10^{-8}$	$1.65 \cdot 10^{-8}$	$1.00 \cdot 10^{-8}$

* Derived from information in Falkowski et al. 1998

of PCB-101 and 153. Considering that more than these five PCB congeners contribute to the total PCB loss rate, the model estimate is in the order of 100 to 1000 tonnes·year^{-1}, i.e. it agrees best with that derived by the method for Estimate 1.

The model results are lower than those derived from method 2, despite the fact that both are based on the same global organic carbon settling rates by Falkowski et al. (1998). This can explained by the fact that the model-calculated concentrations on particles are lower than the global average of 100 ng·g^{-1} organic carbon assumed in method. The calculated global average for the sum of five PCBs was 13 ng·g^{-1}organic carbon in 1970 (range: 3 ng·g^{-1} organic carbon in the Southern hemisphere to 93 ng·g^{-1} organic carbon in northern temperate oceans) and 1 ng·g^{-1} organic carbon in 1999 (range: 0.2 ng·g^{-1} organic carbon in the southern hemisphere to 6 ng·g^{-1} organic carbon in northern temperate oceans). This indicates that method 2 may overestimate the PCB loss by assuming a constant contaminant concentration throughout the global oceans. The model suggest that concentration in the range of 100 ng·g^{-1} organic carbon may indeed be occurring in northern temperate oceans, whereas concentrations are likely much lower in tropical, high latitude and southern hemispheric oceans.

Conclusions

Based on the preceding estimates of the global loss of PCBs we may conclude:

1. The losses of PCBs by atmospheric degradation and loss to the deep sea calculated by the global distribution model are on the same order of magnitude as the semi-empirical estimates based on measured PCB concentrations and fluxes. This increases our confidence in the appropriateness of the process descriptions in the model.
2. There is no reason to doubt the applicability of the reaction rates with OH radicals derived in the laboratory for estimating the real atmospheric lifetimes of POPs. Such lifetimes are longer than may be expected from a cursory inspection of laboratory-derived reaction rates, as a result of low temperatures, variable OH radical concentrations and partitioning of semi-volatile chemicals onto aerosols. Approaches aimed at assessing the long-range transport potential of POPs or candidate POPs need to take this into account (Beyer et al. 2000).
3. Even a chemical with a seemingly fast atmospheric degradation rate can be subject to long-range transport, especially if the chemical is semi-volatile and the atmosphere is cold and/or dark. At higher altitudes, significant partitioning onto particles may limit the loss of semi-volatile organic chemicals by reaction with OH radicals.
4. Models trying to describe the long-range transport behaviour of PCBs need to account for the variability of OH radical concentration, temperature and the latter's influence on the gas/particle partitioning equilibrium.
5. Improving estimations of the atmospheric half-life of PCBs depends primarily on improved quantitative knowledge of (a) the reactions that PCBs may undergo when sorbed to atmospheric particles (especially the kinetics of such reactions), and (b) the vapour/particle partitioning of PCBs at the conditions (low temperatures and pressures, different particle size, composition and concentration) of the higher atmosphere. Other factors which may require refinement are the spatial (vertical and latitudinal) and temporal variability of the concentrations of OH radicals, aerosols and PCB congeners.
6. Atmospheric lifetimes due to reaction with OH radicals for the PCBs in the entire global atmosphere likely range from about 1 month for the light PCBs to in excess of 1 year for the heavy congeners. Lifetimes for PCB mixtures typically present in the atmosphere are likely in the range of approximately 3 months. The global loss of PCBs by reaction with OH radicals during the past 30 years is likely in the range of hundreds to thousands of tonnes per year, most of it due to reactions of the lighter congeners.
7. The global loss of PCBs by settling with organic particles to the deep sea is very likely on the same order of magnitude as the degradation in the atmosphere, but affects the more highly chlorinated congeners more than the lighter ones. Improving estimates of this loss process relies on more meticulous direct measurements of PCB fluxes to the deep sea and PCB concentration in settling particulate matter, in particular in the southern hemisphere. Measurements of the PCB content in deep sea sediment may assist in constraining these estimates.
8. The fact that both global loss processes act to a variable extent on PCBs of different degree of chlorination would suggest that the composition of the PCBs remaining in the global environment should change in time (unless it is influenced strongly by new emissions). A careful analysis of compositional shifts of PCB mixtures observed over a time scale of decades, if they indeed occur, may provide information on the relative and absolute importance of these global loss processes.

9. To estimate global clearance rates from the loss estimates presented in this report, these estimates need to be complemented by estimates of the total inventory of PCBs in the global environment.

Estimated Project Completion Date:

July 2002

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Monitoring the Health of Arctic Peoples and Ecosystems and the Effectiveness of International Controls

Part B: Biotic Monitoring

Retrospective Survey of Organochlorines and Mercury in Arctic Seabird Eggs

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Abstract

Concentrations of total mercury, selenium and a suite of organochlorine compounds were measured in eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*) and black-legged kittiwakes (*Rissa tridactyla*) collected on Prince Leopold Island in Lancaster Sound, Nunavut, between 1975 and 1998. Mercury levels in thick-billed murres and northern fulmar eggs increased significantly during this period whereas concentrations of ΣPCBs (polychlorinated biphenyls), ΣDDT and ΣCBz (chlorinated benzenes) decreased over time for all three species. ΣCHL (chlorodanes) and Σmirex levels decreased in kittiwake eggs while no significant trends were observed for the other two species. Dieldrin concentrations did not change significantly over time for any of the species. Increases in ΣHCH (hexachlorocyclohexane) levels were detected in thick-billed murre eggs but not in northern fulmar and black-legged kittiwake eggs. Levels of the β-HCH isomer, however, increased significantly in murres and fulmars. Stable nitrogen isotope ($\delta^{15}N$) analyses indicate that the temporal trends observed for contaminant concentrations in eggs were not the result of shifts in trophic level. Changing deposition patterns of xenobiotic compounds over the summer and winter ranges of these birds provide a likely explanation for differing exposures through time.

Objectives

To address the question of the utility of Arctic seabird eggs as an indicator of contaminant temporal trends in the Arctic marine environment through the following:

1. reanalysis of archived Arctic seabird egg contents for organochlorines according to a standardized pooling and analytical protocol to confirm whether those residues have been decreasing since the mid-1970s;
2. analysis of archived Arctic seabird egg contents for mercury (Hg) and selenium (Se) to determine whether those levels have been increasing or decreasing since the mid-1970s; and
3. analysis of egg contents and adult livers by mass spectrometry full scan and inductively coupled plasma-mass spectrometry (ICP-MS) to identify any "new" or previously unidentified organochlorines or

metals, respectively, which may have entered the Canadian Arctic food chain.

Introduction

Eggs of seabirds have been used to monitor contamination of the Canadian Arctic marine environment since 1975 (Noble and Elliot 1986, Nettleship and Peakall 1987, Noble 1990, Muir et al. 1999). At the time of egg formation, organochlorine compounds are transferred along with fat to the eggs (Mineau et al. 1984), thus reflecting the contaminant burden in the female at the time of laying (Braune and Norstrom 1989). Both Hg (Fimreite 1979, Lewis and Furness 1993) and Se (Heinz 1996) are also transferred to the eggs. Contaminant burden is comprised of residues assimilated over a long time period and, particularly in migratory species, may represent exposure from a number of different locations (Hebert 1998, Monteiro et al. 1999).

Biomagnification of many chemical residues has been shown to result in higher concentrations of many compounds with increasing trophic level (Riisgard and Hansen 1990, Jarman et al. 1996, Kidd et al. 1998). Therefore, interpretation of contaminant concentrations in seabirds may be confounded if seabird populations shift their dietary patterns over trophic levels through time. Fortunately, retrospective analysis of trophic level in seabirds is possible through the measurement of naturally occurring stable nitrogen isotopes ($\delta^{15}N$) and, to a lesser extent, carbon isotopes ($\delta^{13}C$) (Hobson and Welch 1992, Hobson et al. 1994, Hebert et al. 1999). This is also true of seabird eggs since stable isotope ratios in egg material are expected to reflect the diet of the female prior to, or during, laying (Hobson 1995, Hebert et al. 1999).

Few long-term data sets exist to give a clear picture of how concentrations of contaminants have changed over time at northern latitudes. In the Canadian Arctic, previous contaminant trend information is available for seabird eggs collected on Prince Leopold Island in Lancaster Sound, Nunavut. Contaminants data from samples collected in the mid-1970s (Nettleship and Peakall 1987) have been compared with data from eggs collected in the late 1980s (Noble 1990) and early 1990s (Muir et al. 1999), all of which suggested that levels of many organochlorine compounds were declining. Those samples, however, were analyzed at different times using different analytical techniques. As well, time trends for metal concentrations are nonexistent because concentrations of trace metals were never measured in those samples.

In this study, we provide new insight into contaminant trends in seabirds from the high Arctic by reanalyzing archived samples using standardized laboratory protocols and by extending the trend analyses of eggs from earlier studies. The data for Hg and Se have already been presented (Braune 1999).

Activities in 1999-2000

The organochlorine and stable isotope data resulting from the retrospective analyses of archived egg samples of thick-billed murres (*Uria lomvia*), black-legged kittiwakes (*Rissa tridactyla*) and northern fulmars (*Fulmarus glacialis*) from Prince Leopold Island (74°02' N, 90°05' W) collected from 1975 to 1998 were statistically analyzed, and a manuscript on temporal trends of Hg and organochlorines has been submitted for publication in the journal *Environmental Pollution*. Liver samples of all three species from 1975 and 1993, as well as egg samples from 1993, were analyzed for PCDDs (polychlorodibenzo-*p*-dioxins), PCDFs (polychlorodibenzofurans) and non-ortho PCBs (polychlorinated biphenyls). Those samples have also been submitted for ICP-MS and mass spectrometry full scan analyses. Health Canada's evaluation of risk to human consumers of seabird eggs from Prince Leopold Island and Coats Island (northern Hudson Bay) was completed and the results were sent to the Nunavut Environmental Contaminants Committee.

Results

With the exception of ΣHCH, organochlorine compounds showed declines or, in some cases, no significant change in levels between 1975 and 1998 (Table 1). Levels of ΣPCBs and ΣDDT as well as ΣCBz decreased significantly in eggs of all three species. Concentrations of ΣCHL and Σmirex decreased significantly in black-legged kittiwake eggs but not in the eggs of the other two species. No temporal trends were found for Dieldrin in eggs of any of the three species. Levels of ΣHCH, however, increased approximately two-fold in thick-billed murre eggs between the mid-1970s and the 1990s. No trends in ΣHCH levels were detected in black-legged kittiwake and northern fulmar eggs. The significant increase in ΣHCH in thick-billed murre eggs was the result an increase in concentrations of β-HCH ($p < 0.00001$, $R^2 = 0.69$). Concentrations of α-HCH and γ-HCH did not change significantly in the murre eggs. There was also a significant increase in concentrations of β-HCH in northern fulmars ($p < 0.017$, $R^2 = 0.18$), but due to the simultaneous decrease of α-HCH concentrations ($p < 0.001$, $R^2 = 0.32$), no significant temporal trend resulted from the analysis of ΣHCH (Table 1).

Table 1. Mean concentrations (mg·kg^{-1} wet weight) of organochlorines in eggs of black-legged kittiwakes, northern fulmars and thick-billed murres collected between 1975 and 1998, and results from linear regression analysis

Species	Year	N (# pools)[¶]	% Lipid	ΣPCBs	ΣDDT	ΣCBz	ΣCHL	Dieldrin	ΣMirex	ΣHCH
BLKI[*]	1975	12 (4)	11.9	1.665	0.244	0.080	0.058	0.014	0.015	0.004
	1976	6 (2)	6.0	2.87	0.420	0.095	0.074	0.012	0.026	0.005
	1987	3 (1)	na[†]	0.512	0.108	0.036	0.039	0.008	0.009	0.004
	1993	15 (5)	8.5	0.331	0.071	0.027	0.034	0.017	0.010	0.005
	1998	15 (5)	9.0	0.280	0.060	0.029	0.032	0.009	0.007	0.005
	p			< 0.00001	< 0.00005	< 0.00001	<0.0002	ns[‡]	<0.03	ns
	R^2			0.83	0.67	0.79	0.60		0.29	
NOFU[**]	1975	15 (5)	11.4	0.807	0.674	0.061	0.109	0.013	0.013	0.003
	1976	12 (4)	11.6	1.017	0.859	0.074	0.144	0.017	0.017	0.002
	1977	15 (5)	9.0	0.607	0.425	0.049	0.095	0.011	0.012	0.002
	1987	6 (2)	10.0	0.340	0.218	0.033	0.089	0.011	0.009	0.002
	1993	15 (5)	11.8	0.511	0.396	0.054	0.128	0.017	0.018	0.003
	1998	15 (5)	11.3	0.268	0.209	0.035	0.096	0.014	0.013	0.001
	p			< 0.00001	< 0.00001	< 0.002	ns	ns	ns	ns
	R^2			0.64	0.56	0.30				
TBMU[***]	1975	9 (3)	10.2	0.249	0.197	0.118	0.023	0.014	0.003	0.009
	1976	9 (3)	12.4	0.360	0.232	0.142	0.036	0.028	0.004	0.012
	1977	9 (3)	11.7	0.346	0.232	0.117	0.026	0.012	0.004	0.011
	1987	9 (3)	11.4	0.210	0.156	0.085	0.033	0.017	0.003	0.019
	1988	9 (3)	10.8	0.167	0.104	0.085	0.033	0.016	0.002	0.013
	1993	15 (5)	12.2	0.148	0.139	0.054	0.024	0.013	0.003	0.020
	1998	15 (5)	12.9	0.130	0.100	0.053	0.030	0.015	0.003	0.017
	p			< 0.00001	< 0.00001	< 0.00001	ns	ns	ns	<0.0001
	R^2			0.68	0.60	0.74				0.47

[¶] Total number of birds (number of pools analyzed)

[*] Black-legged kittiwake

[**] Northern fulmar

[***] Thick-billed murre

[†] Not analyzed

[‡] Not significant

Discussion and Conclusions

This study has confirmed decreasing or stable levels of most organochlorine compounds in high Arctic seabirds between 1975 and 1998. However, there has been an increase in concentrations of ΣHCH particularly in the thick-billed murre. Stable isotope ratios in seabird eggs were used as a means to track the trophic status of laying females (Hobson and Welch 1992, Hobson et al. 1994, Jarman et al. 1996, Hebert et al. 1999). Virtually no significant changes in egg isotope ratios were observed for the three species of seabirds throughout this period, suggesting that significant changes in stable isotope values of marine primary producers did not occur and that species did not significantly change trophic positions through time. Therefore, changes in contaminant levels in the eggs appear unrelated to dietary shifts over time.

The significant declines in concentrations of ΣPCBs and ΣDDT in this study corroborate and extend trends based on earlier data reported (Noble 1990, Muir et al. 1999), and are similar to those noted for a number of North

American and Arctic seabirds including those from the Baltic Sea (Olsson and Reutergårdh 1986, Andersson et al. 1988, Bignert et al. 1995), the Barents Sea (Barrett et al. 1996) and the Great Lakes (Hebert et al. 1997). Similar trends for ΣPCBs and ΣDDT have also been found in ringed seals from the Canadian Arctic (Addison and Smith 1998). In most cases, decreasing concentrations in eggs over time reflect declines associated with severe restrictions on the use of these organochlorines in North America since the 1970s. The only organochlorine compounds for which significant increases in concentrations were observed was for ΣHCH, particularly for β-HCH in murres and fulmars, although measured concentrations were quite low. HCH is one of the few organochlorine insecticides still in wide use (Voldner and Li 1995). High volatility of HCHs permits easy partitioning into the atmosphere and transportation via air currents to the Canadian Arctic where HCHs are the most abundant of all organochlorine compounds found in atmospheric samples (Barrie et al. 1997).

Levels of organochlorine compounds in egg samples collected during this study between 1975 and 1998 were below known effect concentrations for eggs of wild birds (see reviews by Blus 1996, Hoffman et al. 1996, Peakall 1996, Wiemeyer 1996). Despite little evidence to indicate an immediate toxic threat from the concentrations of ΣHCH reported in this study, a statistically significant increase in β-HCH levels in murre eggs over time warrants continued monitoring because this isomer is thought to have estrogenic effects (Willett et al. 1998).

Expected Project Completion Date

This is part on an ongoing monitoring program. The next sampling period will be in 2003. A comparison of other available data sets monitoring temporal trends of organochlorines in Arctic biota with the seabird data will be undertaken in 2000-2001.

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Role of Contaminants in Seaduck Population Decline: Metals in Oldsquaw

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Abstract

Western populations of oldsquaw (*Clangula hyemalis*) are in decline and although populations appear stable in the east, oldsquaw are considered to be a *Species of Continental Conservation Concern*. Analyses of metal residues in breast muscle from oldsquaw collected from across the Canadian Arctic during 1991-1994 suggest that western Arctic populations generally have higher levels of cadmium and selenium than eastern populations. Heavy metal contamination from benthic organisms they eat may be an important threat to oldsquaw. Analyses of metal levels in target organs will generate the data necessary to compare with known critical thresholds published in the literature. It is not yet known whether the birds are more exposed to metal contamination on the Arctic breeding grounds or on their overwintering grounds. Measurements of naturally occurring stable isotopes in food webs can, however, be used to discriminate birds that winter in marine areas from those that winter in freshwater environments, such as the Great Lakes. Analyses of metal residue data in combination with stable isotope data are being used to determine if birds overwintering on the Great Lakes risk greater exposure to contaminants than those overwintering in marine environments.

Objectives

To determine the role of contaminants in declining populations of seaducks through the following analyses:

1. archived samples of oldsquaw collected from their Canadian Arctic breeding grounds will be analyzed for mercury (Hg), selenium (Se) and copper (Cu) (in liver); cadmium (Cd) (in kidney); lead (Pb) (in wing bone); and selected samples will be screened for a wider range of metals (in liver); and
2. archived samples of oldsquaw wing bone will be analyzed for stable isotopes ($^{13}C/^{12}C$; $^{15}N/^{14}N$, $^{34}S/^{32}S$) and strontium (Sr) to determine whether birds from certain geographical areas of the Arctic are overwintering in freshwater (i.e. Great Lakes) or marine environments.

Introduction

North American populations of oldsquaw (*Clangula hyemalis*) are declining in the west, but are apparently stable in the east (Canadian Wildlife Service et al. 1997). Nevertheless, oldsquaw are considered to be a *Species of Continental Conservation Concern*. This species nests in greater numbers in the high Arctic than any other duck (Bellrose 1980). Oldsquaw nesting in the eastern Canadian Arctic likely winter along the Atlantic coast whereas western Arctic birds winter in the Bering Sea and along the Pacific coast of Alaska and British Columbia (Bellrose 1980). Considerable numbers also winter on the Great Lakes. It is unknown where the dividing line occurs between the two breeding populations.

Oldsquaw are harvested by Aboriginal people and sport hunters alike, but numbers taken are unlikely to be the cause of the decline (Canadian Wildlife Service et al. 1997). Heavy metal contamination from benthic organisms they eat may be an important threat to oldsquaw. As well, high Pb levels, resulting from spent shot ingested on the breeding grounds, are suspected of impairing reproductive success of oldsquaw nesting in the Alaskan Y-K Delta (Flint et al. 1997). Analyses of metal levels in target organs would generate the data necessary to compare these levels with known critical thresholds published in the literature.

Measurements of naturally occurring stable isotopes in food webs can be used to delineate relative inputs from freshwater and marine biomes (reviewed by Hobson et al. 1997). This is based on the fact that stable isotopes of carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$), sulphur ($\delta^{34}S$) and hydrogen (δD) are typically enriched in marine compared to terrestrial or freshwater systems and these isotopic signatures are passed on to the consumers. Upon arrival on the breeding grounds, metabolically active tissues of birds that wintered in freshwater environments can be distinguished isotopically from those that wintered in marine areas. This approach can also be applied to eggs if birds mobilize endogenous reserves to eggs shortly after arrival on the breeding grounds (Hobson et al. 1997). Such information can be used to determine if birds overwintering on the Great Lakes risk greater exposure to contaminants than those overwintering in marine environments.

Metal residue data for breast muscle from oldsquaw collected from across the Canadian Arctic during 1991-1994 suggest that western Arctic populations generally have higher levels of Cd and Se than eastern populations (Braune et al. 1999). It is not known whether the birds are more exposed to metal contamination on the Arctic breeding grounds or on their overwintering grounds. The stable isotope measurements should discriminate those birds that overwinter on the Great Lakes, a known contaminated area. As well, Sr measurements should help to corroborate stable isotope findings since Sr levels are known to be higher in marine than freshwater systems. It should then be possible to determine if those birds overwintering in the Great Lakes also contain higher tissue residue levels than the others. The connection between risk of exposure and overwintering area can then be investigated, if such a link exists. Alternatively, if no association between metal levels and overwintering area can be established, the source of contamination may, in fact, be in the Arctic.

Activities in 1999-2000

The tissues of 101 adult oldsquaw from the Canadian Arctic are archived in the Canadian Wildlife Service (CWS) Specimen Bank: 59 birds from the eastern Arctic (east of 95°W) and 42 birds from the western Arctic. Liver, kidney and bone samples have been retrieved from the CWS Specimen Bank for each of the 101 birds and have been submitted for metal analyses (Hg, Se, Cu in liver; Cd in kidney; Pb in bone). Livers of 10 birds with elevated levels of metals are to be analyzed by inductively coupled plasma-mass spectrometry to screen for levels of other metals, but selection of those samples is pending review of the analytical results for the 101 birds.

Results

Results for the metal analyses are not yet available.

Discussion and Conclusions

Bone samples from the same 101 birds for which metals are being analyzed will be analyzed for stable isotopes as well as Sr to discriminate between ducks wintering in freshwater or marine environments. Stable isotope values in bird tissues are in dynamic equilibrium with local food webs; therefore, it is necessary to choose a tissue with slow turnover rate to delineate between freshwater and marine wintering ducks. Bone collagen was chosen because it has a turnover rate slow enough for isotope measurements to represent lifetime average values (Hobson and Sealy 1991). Sr will be analyzed in addition to stable isotopes in bone collagen to help discriminate between ducks that winter in freshwater and those that winter in marine environments. All chemical and stable isotope analyses should be completed in 2000-2001, and results will be reported in 2001-2002.

Expected Project Completion Date

March 31, 2002

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Community-Based Monitoring of Abnormalities in Wildlife

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Abstract

Communities across the North have a long history of dependence on wildlife and the environment. Changes in wildlife health can have significant impacts on individual animal and population health, as well as on human use of the resource. Although most wild animals are healthy, abnormalities such as diseases, parasites, tumours and deformities can occur in any wildlife population. Hunters and elders have reported an increase in observed changes in wildlife health in recent times, and have questioned if contaminants may have a role in these changes or if they are naturally occurring. Documenting the occurrence of these changes is the first step in addressing these questions, and may serve as a sentinel to identify problems requiring further investigation or to develop testable hypotheses. Through their land-based subsistence lifestyles, local hunters have developed a wealth of experience and understanding of the ecosystems in which they live, and can recognize changes in animals and trends in the occurrence of some conditions over time. This 3-year pilot project designed a system for collecting and organizing information from hunters and elders on unusual changes that may have been noticed in wildlife populations. This included the development of a community-based system to systematically collect and investigate hunter observations and/or samples of abnormalities in wildlife, and a Traditional Ecological Knowledge survey of disease and/or abnormalities in wildlife. Results of the pilot project suggest that a community-based monitoring system can be an effective approach to monitor selected grossly visible abnormalities, most of which are naturally occurring conditions.

Objectives

1. To develop a community-based monitoring program in which harvesters can document and communicate observations of changes in wildlife in a systematic and useful way.
2. To provide an "early warning system" to detect changes or patterns in wildlife health at an ecosystem level. This systematic monitoring may identify areas

requiring further study and aid in hypothesis development.

3. To integrate scientific and traditional ecological knowledge to increase general understanding of changes in the health status of wildlife.
4. To allow communities to participate and build local capacity to identify, investigate and respond to changes in the wildlife resources they harvest.

Introduction

Communities across the North have a long history of dependence on wildlife and the environment. The traditional subsistence harvesting of fish, terrestrial wildlife, marine mammals and migratory birds provides northerners with significant nutritional, economic, social and cultural benefits. Traditional pursuits such as hunting, fishing and trapping have long been the basis of the northern economy. The replacement value of country food and other renewable resources is approximately $60 million per year (1990 dollars). Confidence in the health of wildlife populations and the availability of safe and healthy meat is extremely important for maintaining both subsistence harvesting and the commercial use of wildlife.

Changes in wildlife health can have significant impacts on individual animal and population health, as well as human use of the resource. Although most wild animals are healthy, abnormalities such as diseases, parasites, tumours and deformities can occur in any wildlife population. Some conditions appear to be increasing in frequency or appearing in new areas of the North. Local people want to know if there is a connection between the environmental contaminants they are being told about and their observations of unusual changes or abnormalities in wildlife. Documenting the occurrence of these changes is the first step in addressing this question. Answers to this question may serve as a sentinels for identifying problems requiring further investigation. Such documentation may also focus attention on linkages between observed changes and known effects of specific contaminants, and assist in developing testable hypotheses.

Information on abnormalities in wildlife has typically been obtained in several ways. There have been a number of specific studies focussing on wildlife health in individual species. Changes have also been detected opportunistically when handling animals during other wildlife studies or commercial wildlife harvests. Samples or reports of abnormalities are also periodically brought in to Renewable Resource Officers by hunters who come across unusual findings, but this system is primarily a reactive process to individual cases. Under this system, the number of samples or observations received has been low compared to the number of animals harvested, the expertise of the harvesters has not been fully accessed, and it has been difficult to differentiate localized versus widespread patterns.

Through their land-based subsistence lifestyles, local hunters have developed a wealth of experience and understanding of the ecosystems in which they live. Experienced hunters spend large amounts of time observing wildlife, and can recognize changes in animals and trends in the occurrence of some conditions over time. This field of ecological knowledge is commonly known as Traditional Ecological Knowledge (TEK), and includes empirical knowledge gained through experience and observation. This is consistent with Western science, where experience and observation form the basis of modern empirical research. The scientific process differs in that it has developed a formal method by which these experiences and observations can be systematically documented and verified. The opportunity exists to integrate these two systems to increase information on changes in wildlife health, and to increase trust and co-operation between resource harvesters and researchers.

Activities in 1999-2000

This pilot project was conducted over a 3-year period ending in 1999-2000. At the beginning of the project, an emphasis was placed on capacity building in the communities through local training sessions and the development of resource materials. During the later stages of the project, the emphasis was placed on community-based monitoring of abnormalities through both passive and active surveillance activities. Training and updating of resource material continued throughout the duration of the project.

Local training sessions

Over the life of the program, a wide range of training and information sessions were held in each of the three participating communities. As the emphasis was placed on community participation, these sessions targeted a wide range of community groups including local resource harvesters (hunters and trappers), food preparers, elders, students, local environmental committees and interested members of the general public. These sessions were designed to introduce the project, provide background information on wildlife health, and begin to build local capacity to manage the project and respond to changes in the wildlife resources they harvest. The goal of the training component of this work was to build capacity within the community to deal with abnormalities in a timely fashion. While training was emphasized at the beginning of the program, additional training sessions were provided throughout the life of the project. In addition to community-based workshops and meetings, field training courses were held in Fort Resolution and Kugluktuk. Local hunters, elders and students participated in hands-on training in the field, and applying information and techniques from the training sessions on harvested animals in the field.

Development of resource material

A self-contained package of resource materials was developed to assist with implementing the project. This package was used by resource harvesters to assist with uniform collection and recording of information, sample collection and testing, and interpretation and reporting of results. The resource kit included a project manual, a hunter/elder survey form, standardized disease observation and sample submission forms, visual (photographic) reference guides to assist with identification of abnormalities, and field sampling kits. These materials were field tested as part of the community-based monitoring program, and were updated and reprinted in the final year of the program. A *Field Guide to Common Diseases and Parasites of Wildlife in the NWT and Nunavut* (initially produced in 1998-1999; reprinted in 1999-2000) was particularly well received in communities as a tool for hunters to identify abnormalities and make timely decisions on the implications for use of the carcass.

Community-based monitoring of abnormalities in wildlife

Community-based monitoring of abnormalities in wildlife began in 1997-1998, and continued in 1998-1999 and 1999-2000. Various approaches were considered to maximize the effectiveness of sample collection and the documentation of observations from wildlife harvesters within each community. Information was collected on the occurrence of abnormalities in wildlife from all local hunters using written reports and/or samples for analysis, with an emphasis on key harvesters in the community. Several field collections of caribou from herds harvested by all three communities were also conducted to provide baseline data on their health status, and to provide a method to validate data collected by the community-based monitoring system using hunter submitted samples. TEK surveys on the occurrence of abnormalities in wildlife were also conducted with elders and experienced wildlife harvesters in each community. Data from the community-based monitoring system and systematic health surveys are currently being analyzed and interpreted.

TEK survey

Interviews were conducted with experienced, long-time local harvesters about the occurrence of abnormalities and diseases in wildlife. The community coordinators or alternate local wildlife harvesters conducted the interviews. This eliminated the need for simultaneous translation, ensured the interviewer was familiar with the topic, and enabled elders to relax and share their knowledge of the subject. Surveys were conducted in Fort Resolution, Fort Good Hope and Kugluktuk; results are currently being evaluated.

Results and Discussion

This initiative was conducted as a pilot project to test the concept, design and implementation of a community-based monitoring system in three northern communities. The communities were selected to reflect regional differences in wildlife species and harvesting patterns: Kugluktuk, Nunavut (Arctic Coast), Fort Good Hope, Northwest Territories (Mackenzie Valley) and Fort Resolution, Northwest Territories (Slave River Delta). As a community-based process, a local coordinator was identified in each community. This individual helped organize community wildlife health/sampling workshops, routinely talked with local hunters to document abnormalities in harvested wildlife, collected samples of abnormalities from hunters and submitted them for testing, facilitated result reporting, and organized traditional knowledge surveys of respected local hunters about abnormalities in wildlife. The system was designed to rely on key observers or monitors in each community who are recognized for their wildlife harvesting and observation skills.

Community-based monitoring of abnormalities was conducted over the 3 years of the pilot project. Documentation of observations by wildlife harvesters was done through both sample and report collection from local hunters and selected key harvesters in the community. This provided information on both the types and proximate cause of abnormalities being seen by harvesters, and an estimate of frequency of their occurrence. A wide range of abnormalities were reported by hunters and trappers, with the vast majority being "naturally occurring" diseases and parasites that occur in northern wildlife species. The frequently occurring conditions include brucellosis, tuberculosis, warbles (*Oedemagena tarandi*), liver parasites (*Taenia hydatigena*), muscle parasites (*Sarcocystis* spp. and *Taenia krabbei*), gastrointestinal parasites (*Nematodirus* spp., *Moniezia* spp., *Eimeria* spp.), lung worms (*Dictyocaulus* spp.), hydatid disease (*Echinococcus* *granulosus*), ticks (*Dermacentor albipictus*), physical injuries and malnutrition. Very few tumours or developmental anomalies have been detected. Final analysis and interpretation of the data is currently being completed.

As a method to validate data collected under the community-based monitoring system using hunter submitted samples, several systematic disease surveys were conducted during the same time period to compare the data on type and prevalence of abnormalities provided by both systems. Three field collections of caribou were conducted to provide baseline data on the health status and disease and/or parasite prevalence in caribou from herds being harvested by Kugluktuk, Fort Resolution and Fort Good Hope hunters. Collections were held in both fall and spring to look at seasonal changes in the prevalence of abnormalities and diseases at times when caribou are normally harvested. A total of 77 caribou taken during three separate collections were tested for both grossly visible and subclinical conditions. The animals collected were generally very healthy, with a low prevalence of diseases and parasites. As in the community-based monitoring, primarily "naturally occurring" conditions were noted with a very low prevalence of neoplasia or developmental anomalies. The most common diseases and parasites detected and their seasonal prevalence are listed in Table 1.

Overall, both the type and relative prevalence of diseases and parasites detected were generally comparable between the community-based monitoring program and the systematic disease survey. The one major discrepancy between the two monitoring systems occurred in the detection of the few diseases or parasites that do not produce grossly visible lesions consistently in infected animals. This was particularly evident with the parasite *Sarcocystis*, which has a very high prevalence in the population but only occasionally produces clinical signs in caribou that are visible to hunters. Overall, however, these preliminary results are encouraging for the use of a community-based monitoring system for the detection of grossly visible conditions. This type of system appears to have promise as a local, effective method of monitoring abnormalities in wildlife.

Project Completion Date

March 31, 2000

Table 1. Diseases and parasites detected in caribou from herds harvested by three project communities

Condition	Bluenose (Fall) (n = 26)	Bluenose (Spring) (n = 26)	Bathurst (Spring) (n = 25)
Body condition (mean back fat depth)	11.8 mm	5.4 mm	20.4 mm
Brucellosis (*Brucella suis*; biovar 4)	7.7%	4.0%	0%
Warbles (*Oedemagena tarandi*)	44.4% (range: 3-145/animal)	100% (range: 8-182/animal)	100% (range: 7-126/animal)
Sarcocystosis (*Sarcocystis* spp.)	92.3% (67% mild, 29% moderate, 4% severe)	100% (92% mild, 8% moderate, 0% severe)	results pending
Besnoitiosis (*Besnoitia* spp.)	46 % (1 clinical case; 11 positive but not visible)	results pending	results pending
Liver tapeworms (*Taenia hydatigena*)	11.5% (3 cases)	0% (0 cases)	44.0% (11 cases)
Muscle tapeworms (*Taenia krabbei*)	0%	0%	20.0% (5 cases)
Gastrointestinal parasites (*Nematodirus*, *Moniezia* and *Eimeria* spp.)	15.4% (4 cases)	26.9% (7 cases)	12.0% (3 cases)
Lung worms (*Protostrongylus* spp.; larvae)	19.2% (range: 42-544/g)	38.5% (range: 1-125/g)	36.0% (range: 1-331/g)
Lung worms (*Dictyocaulus* spp.)	7.7% (2 cases)	results pending	results pending
Non-specific infections	3.8% (1 case)	3.8% (1 case)	0%
Developmental anomalies	3.8%	0%	0%
Tumours/Neoplasia	0%	0%	0%

Metal and Radionuclide Accumulation and Effects in Caribou (*Rangifer tarandus*)

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Abstract

Metals and radionuclides were measured in fourteen female and five male caribou (mean age 5.4 years) from the Lake Harbour herd near Kimmirut, Nunavut, on south Baffin Island. The tissues were collected in March 1999 as part of an ongoing program to monitor the levels of these elements in herds throughout the Northwest Territories and Nunavut and to establish temporal trends from earlier surveys for compounds with anthropogenic sources. Cadmium averaged 26.9 mg·kg^{-1} dry wt in kidney, close to the level of 31.98 mg·kg^{-1} reported in the 1992 survey, and increased significantly ($p < 0.001$) with age. These levels are among the upper range of values observed in Nunavut and the Northwest Territories, but are much lower than those in some Yukon herds living in mineralized areas. Other metals generally correspond with the values reported in the earlier survey. ^{137}Cs, a radionuclide left over from the atmospheric nuclear weapons tests in the 1960s fell to 69.8 Bq·kg^{-1} wet wt in muscle from 111 Bq·kg^{-1} wet wt in 1992, consistent with an environmental half-life of 8 to 10 years. The natural radionuclides ^{210}Pb and ^{210}Po remain high in this herd, with an average ^{210}Pb concentration of 1210 Bq·kg^{-1} in bone, including one individual in excess of 2300 Bq·kg^{-1}. The survey indicates consistent regional differences between herds in terms of metal and radionuclide concentrations and also suggests a need to understand the factors behind those regional differences.

Objectives

1. To verify high levels of metals (cadmium [Cd], aluminum, manganese) and radionuclides (lead-210 [^{210}Pb] and polonium-210 [^{210}Po]) detected in previous studies in caribou.
2. To analyze several tissues for a range of elements and isotopes (uranium, thorium, stable strontium, titanium) which may provide insight into the source of contamination.

3. To contribute information on temporal trends in contaminant exposure in the mainland Bluenose and south Baffin Island caribou herds.

Introduction

This project is part of an ongoing program to assess metal and radionuclide concentrations in barren ground caribou (*Rangifer tarandus groenlandicus*) throughout the Northwest Territories and Nunavut. The program fulfills two crucial roles in the Northern Contaminants Program: i) monitoring contaminant residue levels in important traditional/country food species; and ii) the long-term monitoring of temporal trends. A major survey of contaminants in caribou (Elkin and Bethke 1995) showed that organochlorines in caribou are low relative to marine mammals and some fish species, however metals and radionuclides can vary significantly between regions. Metals, such as Cd, may approach the concentrations in liver and kidney in which consumption advisories may be considered.

This report summarizes the levels of metals and radionuclides in the Lake Harbour herd, which is situated near Kimmirut on south Baffin Island. The herd was selected for analysis as part of a continuing survey of contaminants in caribou and because of high levels of metals and natural radionuclides found in the herd in the 1992-1993 survey (Elkin and Bethke 1995, Macdonald et al. 1996).

Barren-ground caribou are found across the Northwest Territories, and are a major component of the traditional diet in northern communities. Many factors can influence the individual and population health of caribou, and changes in population condition are often multifactorial in origin. Contaminants are one group of stressors that may influence the health status of a herd by altering tissue structure and function, and by acting in association with other stresses to impair reproduction, immune function or other body functions. The need for work on the biological effects of contaminants in caribou was highlighted by the results of earlier studies (Elkin and Bethke 1995, Macdonald et al. 1996) that showed elevated levels of metals and radionuclides in some caribou herds. The concentrations of metals (Cd) and naturally-occurring radionuclides (^{210}Pb and ^{210}Po) in the south Baffin Island caribou herd near Kimmirut (Lake Harbour) were shown to be higher than other herds in the central and western Arctic (Elkin and Bethke 1995, Macdonald et al. 1996). The concentrations were high enough in some individuals that some effects were expected at the tissue level. The high contaminant concentrations, which may be natural, prompted concerns for the health of the caribou and also suggested that the metal levels should be measured again to confirm the results and to establish temporal trends for those compounds with an anthropogenic source.

Activities in 1999-2000

Field sampling

Nineteen caribou were collected in early March 1999 near Kimmirut, Nunavut, on south Baffin Island. The collection was made by Resources, Wildlife & Economic Development personnel and two hunters from Kimmirut, in cooperation with the Mayukalik Hunters and Trappers Association. Sex, body condition and other biological and morphometric data were taken at the time of collection and a central incisor was removed for aging by cementum analysis. Liver, kidney, muscle and bone samples were collected for metal and radionuclide analysis. Blood and tissue samples were collected for general health and disease assessment. Upon completion of the field collection, the caribou meat from the collection was provided to the community for local use.

Metals were analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP/MS), following digestion with nitric acid, by EnviroTest Laboratories in Edmonton, Alberta. These laboratories are Canadian Association for Environmental Analytical Laboratories accredited. Quality assurance and quality control (QA/QC) of the analysis included blanks, spiked samples, standard reference materials (e.g. bovine liver) and duplicates.

A detailed description of the radionuclide analysis is given in Macdonald et al. (1996). Gamma-emitting radionuclides of cesium (^{137}Cs), potassium (^{40}K), radon (^{226}Rn), lead (^{210}Pb) and thorium (^{232}Th) were analyzed at Whiteshell Laboratories using a p-type well detector in tissue ashed at approximately 500°C. ^{210}Po was analyzed by alpha spectrometry using a ^{209}Po tracer, after plating onto silver discs. ^{210}Po concentration at the time of sampling was calculated as the sum of the supported fraction (^{210}Pb concentration) and the unsupported fraction, corrected for decay from counting date to plating date. QA/QC procedures included the use of standard radiation sources, duplicates and blanks.

Results and Discussion

A total of 14 female and 5 males were harvested. Ages ranged from 1 to 10 years, with a pooled mean for both sexes of 5.4 years. Total and dressed weights could not be obtained due to technical reasons. Mean total body length was 172.1 cm (standard deviation [SD] = 9.3) with a chest girth of 109.4 cm (SD = 5.5). Back fat ranged from trace amounts to 20 mm in one 5 year-old female.

Metal concentrations in the liver and kidney are summarized in Table 1. The concentrations in Table 1 are expressed on a wet weight basis using a conversion from dry weight of 20.9% moisture for kidney and 29.3% for liver

Table 1. Mean concentrations of metals (mg·kg^{-1} wet wt) in liver and kidney in Lake Harbour caribou, March 1999*

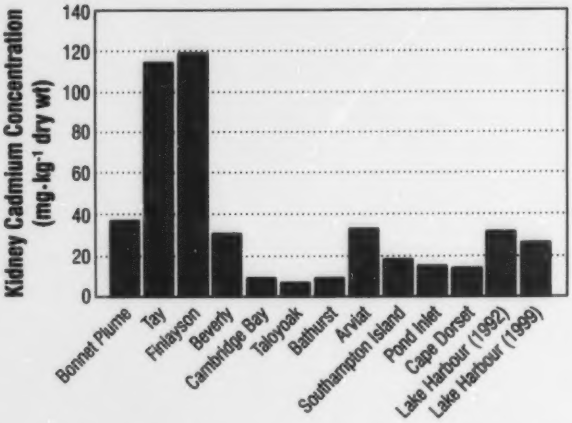
Metal	Liver (n=19)	Kidney (n=19)
Aluminum	2.06 (1.67 - 2.55)	1.66 (1.41 - 1.94)
Barium	0.21 (0.16 - 0.28)	0.32 (0.25 - 0.42)
Boron	0.78 (0.56 - 1.09)	< 0.4
Cadmium	1.02 (0.65 - 1.58)	4.71 (2.45 - 9.07)
Calcium	41.9 (32.4 - 54.2)	87.9 (74.8 - 103)
Chromium	< 0.4	0.18 (0.11 - 0.31)
Cobalt	0.12 (0.10 - 0.14)	0.08 (0.07 - 0.10)
Copper	30.0 (21.5 - 42.0)	3.07 (2.78 - 3.38)
Iron	792 (560 - 1120)	39.2 (31.6 - 48.6)
Lead	2.27 (1.56 - 3.32)	0.38 (0.31 - 0.48)
Magnesium	146 (135 - 157)	134 (127 - 142)
Manganese	2.47 (2.18 - 2.81)	1.98 (1.83 - 2.14)
Molybdenum	0.72 (0.63 - 0.83)	0.11 (0.09 - 0.14)
Nickel	0.09 (0.06 - 0.13)	0.10 (0.08 - 0.14)
Phosphorus	3137 (3013 - 3266)	2384 (2229 - 2550)
Potassium	2522 (2377 - 2674)	2372 (2217 - 2539)
Selenium	0.53 (0.45 - 0.62)	0.91 (0.82 - 1.02)
Sodium	564 (479 - 663)	1159 (1063 - 1264)
Strontium	0.04 (0.04 - 0.05)	0.10 (0.08 - 0.13)
Zinc	18.3 (16.4 - 20.6)	14.5 (12.7 - 16.5)
Mercury	0.22 (0.16 - 0.31)	0.02 (0.01 - 0.03)

* Values are geometric means with geometric standard deviations in brackets.

(Macdonald et al. 1996). Antimony, arsenic, beryllium, silver, thallium, tin, uranium were below detection limits in all samples of both tissues. Chromium and vanadium were not detected in liver and boron was not detected in kidney.

The results for metals generally support earlier data in this herd, and the relative levels of the metals in liver and kidney correspond to data from the Bluenose herd collected in 1998, which were also analyzed by ICP/MS. Cd in kidney has a geometric mean of 4.71 mg·kg^{-1} on a wet weight basis, which corresponds to a mean of 26.9 mg·kg^{-1} on a dry weight basis, close to the value of 31.98 mg·kg^{-1} reported by Elkin and Bethke (1995) in 1992 for this herd. The present collection includes two individuals with Cd levels of 65 mg·kg^{-1}. Cd concentration in kidney increased significantly with age ($p < 0.001$). Other than the very high Cd levels reported for herds in mineralised areas of the Yukon (Gamberg and Scheuhammer 1994), these levels are among the highest levels reported for barren ground caribou in Nunavut and the Northwest Territories. A comparison of Cd levels in kidney from the Yukon, Northwest Territories and Nunavut (Figure 1) shows large variation, with the concentrations in the Lake Harbour herd remaining at about 30 mg·kg^{-1} dry weight over the two collections in 1992 and 1999. The mean Cd value is significantly below the tissue guideline of 30 mg·kg^{-1} wet weight in kidney recommended by Environment Canada, above which biological effects are expected to occur in some species (Canadian Environmental Protection Act 1994).

Figure 1. Cadmium concentration (mg·kg^{-1} dry wt) in caribou kidney collected at several sites in the Yukon, Northwest Territories and Nunavut*



* Source: Yukon Contaminants Committee, Elkin and Bethke (1995) and the present study

Other metals show significant variation with the 1992 collection from Lake Harbour. In the present analysis, the mean iron level in liver was 792 mg·kg^{-1} wet wt, or 2860 mg·kg^{-1} on a dry weight basis, which is substantially lower than the value of almost 4000 mg·kg^{-1} dry wt reported for liver by Elkin and Bethke (1995). Similarly, the level of 29.57 mg·kg^{-1} dry wt reported for copper in kidney in 1992 is markedly higher than the levels of 14.7 mg·kg^{-1} dry wt reported here. Variances may be due to seasonal metabolic changes in the caribou or due to analytical differences in the two collections. A subset of the 1999 tissues are being re-analyzed by the contract lab to determine the reproducibility of the analyses. The levels of mercury in liver are low relative to marine mammals but are equivalent to the levels observed in fish in some areas of the Northwest Territories (0.22 mg·kg^{-1} wet wt or 0.80 mg·kg^{-1} dry wt).

The concentration of the human-made radionuclide ^{137}Cs is substantially lower in muscle in 1999 than in data from collections in 1992 and 1993. Muscle concentrations of ^{137}Cs were 111 Bq·kg^{-1} in 1992 and 127 Bq·kg^{-1} in 1993 (Macdonald et al. 1996), and have declined to 69.8 Bq·kg^{-1}

Table 2. Concentration of radionuclides (Bq·kg^{-1} wet wt) in Lake Harbour caribou collected near Kimmirut in 1999*

Tissue	Moisture Content (% wet wt)	Ash Content (% wet wt)	^{40}K	^{137}Cs	^{210}Pb	^{232}Th	^{226}Ra	^{241}Am	^{235}U	^{210}Po
Muscle	26.9 (1.0)	1.17 (0.05)	187 (140 - 250)	69.8 (53.6 - 91.1)	< 23.8	< 8.0	< 9.0	< 2.4	< 12.0	na**
Liver	31.4 (1.1)	1.57 (0.05)	170 (151 - 193)	35.7 (27.9 - 45.8)	393 (270 - 571)	< 3.02	< 3.56	< 0.99	< 4.54	394 (219 - 711)
Kidney	22.1 (1.2)	1.47 (0.24)	166 (148 - 185)	100 (76.5 - 131)	80.6 (61.7 - 105)	< 0.99	< 1.85	< 0.55	< 2.46	305 (185 - 502)
Spleen	22.0 (0.78)	1.16 (0.07)	191 (178 - 204)	32.0 (24.0 - 42.7)	7.7 (4.7 - 12.6)	< 1.12	< 1.25	< 0.40	< 1.87	192 (141 - 260)
Bone	89.3 (0.64)	65.5 (1.0)	< 82.6	2.83 (1.60 - 5.01)	1210 (855 - 1711)	9.25 (7.20 - 11.9)	7.62 (4.96 - 11.7)	< 2.06	< 5.43	na
Marrow	—	0.44*** (0.08)	—	0.46 (0.32 - 0.67)	5.82 (4.43 - 7.65)	0.35 (0.16 - 0.76)	< 0.80	< 0.18	< 0.91	12.8 (9.13 - 17.9)

* Data are arithmetic means (standard deviation) for moisture and ash content. Radionuclide concentrations are geometric means, with geometric standard deviations in brackets.

** Not analyzed

*** Ash content of marrow expressed as percent of freeze-dried weight

wet wt in 1999. Similar rates of decline were observed in liver and kidney. This decline supports environmental half-life estimates of 8 to 10 years suggested elsewhere for ^{137}Cs (Macdonald et al. 1996), and suggests that ^{137}Cs will continue to be a decreasing problem in the North if there are no new sources of the isotope. There is no evidence of other human-made nuclides, such as americium-241 (^{241}Am), in any of the organs.

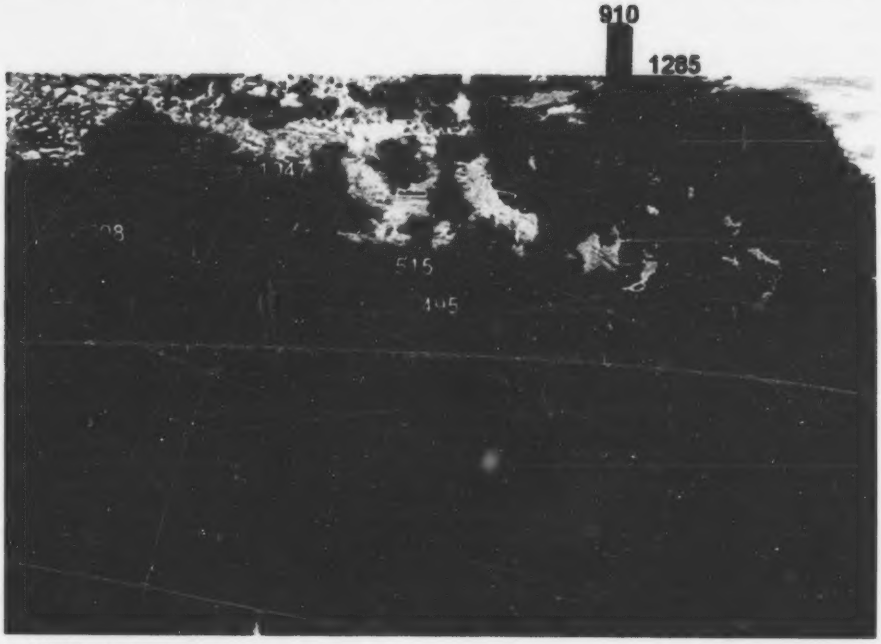
A major component of the present study was to corroborate high levels of natural radionuclides observed in the Lake Harbour herd in the early 1990s. The levels reported here (Table 2) for the naturally-occurring radionuclides ^{210}Pb and ^{210}Po closely correspond to the levels observed in all organs in earlier studies. Macdonald et al. (1996) reported a mean ^{210}Pb level of 1285 Bq·kg^{-1} in bone, which was the highest level observed in 13 herds of caribou distributed across the North. A similar mean of 1210 Bq·kg^{-1} was measured in the 1999 collection, which includes one individual in excess of 2300 Bq·kg^{-1}. ^{210}Po, an alpha-emitting radionuclide which arises from the decay of ^{210}Pb, has a mean concentration of 305 and 394 in kidney and liver, respectively. A comparison of ^{210}Pb levels in bone in several herds shows relatively low levels in the Yukon and northwest Canadian Arctic, but higher levels in the eastern Arctic (Figure 2).

The high levels of ^{210}Pb and ^{210}Po observed in this herd in the 1992 and 1999 collections support the observation that this herd undergoes the highest background radiation dose of caribou in northern Canada, and probably among non-human species in Canada. A recent assessment of dose calculations to non-human species by the Canadian Nuclear Safety Commission (formerly the Atomic Energy Control Board) suggested that the weighting factor for alpha radiation, which includes ^{210}Po, should be increased from the current value of 20 to take into account new data on the effects of alpha radiation in some species. This would place the dose in these caribou at very high levels (approximately 1 Gy·y^{-1}). At present, there is no evidence of major biological effects in this herd. The reasons for the high levels are unknown but could include mineralization which causes a release of uranium from uranium-rich rocks onto lichen or the deposition of ^{210}Pb onto lichens from natural or anthropogenic sources. The Beverly herd, which also shows high ^{210}Pb levels in bone, migrates through areas known to be rich in uranium (i.e. northern Saskatchewan), and the elevated ^{210}Pb is probably due to natural food chain transfer.

Conclusions

These data support the values reported in earlier studies showing elevated concentrations of some inorganic compounds in the Lake Harbour herd on south Baffin Island. Cd levels in kidney are high relative to some other herds, but below the levels expected to be toxic, and well below the maxima seen in some caribou in the Yukon. Elements such as iron, copper and zinc are lower than in other herds, but this may be due to natural fluctuations in these metabolically-regulated metals.

Figure 2. ^{210}Pb concentration in caribou bone taken from surveys conducted in the early 1990s to the present*



* Source: Macdonald et al. (1996) and the present study

^{137}Cs is declining at rates that correspond to estimates of an environmental half-life of 8 to10 years. Background radiation dose is high in this herd due to high levels of ^{210}Pb and ^{210}Po, and the potential effects from this exposure is unknown.

Expected Project Completion Date

Ongoing

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An Investigation of Factors Affecting High Mercury Concentrations in Predatory Fish in the Mackenzie River Basin

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Abstract

This study is investigating why predatory fish such as pike, walleye and lake trout in some lakes in the Mackenzie River Basin have very high concentrations of mercury in their tissues. This is a widespread concern and this study is trying to find out why mercury levels are so high in some lakes and not in others. Research in 1999 focussed on Cli Lake and Little Doctor Lake, two high mercury lakes. A core collected in March 1999 was dated and sedimentation rates measured. Carbon, nitrogen and phosphorus analyses also were conducted; mercury analyses are ongoing. In July, research sampling was again conducted on Cli Lake, with lesser effort on Little Doctor Lake. Mercury occurred in moderate concentrations in lake water and stream sediments. Invertebrates, forage fish, pike and lake trout were collected for stable isotope and mercury analyses. Organic contaminants are being measured in the food web to assess concentrations in organisms consumed by people and biomagnification rates relative to better studied systems such as Great Slave Lake and Yukon lakes.

Objectives

1. To conduct limnological studies to determine why predatory fish inhabiting some lakes in the Mackenzie River Basin have elevated mercury (Hg) levels while fish in other apparently similar lakes have lower Hg levels.
2. Determine the degree to which Hg flux rates have increased in high- and low-Hg lakes. Assess the reasons why these increases may have occurred. Also determine whether Hg flux rates are higher in lakes in which fish have high Hg body burdens than in lakes in which fish have lower Hg body burdens.

3. Investigate the role of food web structure in the biomagnification of Hg by predatory fish. Investigate whether there are differences in food web structure in high- and low-Hg lakes.
4. Investigate whether it is possible to develop lake-specific remedial actions to reduce Hg concentrations in harvested fish from high-Hg lakes, e.g. selective fishing (Verta 1990).
5. As part of the Hg studies, conduct limited determinations of persistent organic pollutants (POPs) concentrations in predatory fish to assess whether contaminant levels are below human health consumption guidelines. This POPs study also will allow us to compare Hg and organochlorine biomagnification rates.

Introduction

Fish are an important natural resource for communities living in the Mackenzie River Valley. Fish have been and will continue to be important in subsistence fisheries and in maintaining cultural values in the North (Evans and Carpenter 1997). Moreover, there is a growing interest in the increased use of fish for commercial purposes, both for domestic sale and for the further development of tourist-based sports fisheries. For these reasons, the Department of Fisheries and Oceans (DFO) has been working with communities in conducting stock assessment studies of these fisheries. Hg levels have routinely been determined in fish as part of the DFO Inspection Service program (Jensen et al. 1997).

These studies have revealed that there are large spatial variations in Hg concentrations in predatory fish in the Northwest Territories (NWT), particularly in the Mackenzie River Basin. Moreover, in many instances, Hg levels exceed both the 0.2 $\mu g \cdot g^{-1}$ recommended guideline for frequent consumers of fish and 0.5 $\mu g \cdot g^{-1}$, the upper limit for the commercial sale of fish. Examples include Cli Lake, Little Doctor Lake, Lac-à-Jacques, Turton Lake and Lac-Ste-Thérèse (Lyle Lockhart, personal communication). Hg levels are lower than in other lakes such as Lac Belot and Great Bear Lake. It is not known why Hg levels are very high in predatory fish inhabiting some lakes but not in other lakes. As a consequence, it is not known what actions can be taken to reduce environmental concerns related to Hg (and possibly to other metals) in the environment.

Our project is investigating why predatory fish in some lakes in the Mackenzie River Basin have very high concentrations of Hg in their tissue while fish in other lakes have low Hg concentrations. This project is linked with Lockhart and Evans' Northern Contaminants Program (NCP) project, Mercury in Fish from Stock Surveys in Lakes in the Western Northwest Territories. Furthermore, because these lakes are largely unstudied, our study is obtaining fundamental information on the limnology and fisheries of selected high-Hg and low-Hg lakes. By obtaining these baseline limnological and fisheries data, we will be better able to investigate the factors affecting high Hg concentrations in predatory fish. The overall goal of the study is to determine whether or not remedial actions can be developed to reduce this human health and environmental concern.

Activities in 1999-2000

In 1999, we continued our research focussing on Cli Lake, a high-Hg lake and the site of a naturalist lodge operated by community members from Fort Simpson. Lesser effort was focussed on Little Doctor Lake; this lake does not have the same infrastructure in place for scientific sampling as Cli Lake. Highlights of the studies are discussed below.

Results and Discussion

Two cores were collected from Cli Lake in March 1999. This core had an estimated sedimentation rate of 166 $g \cdot m^{-2} \cdot yr^{-1}$, a low rate that was further reduced to 80 $g \cdot m^{-2} \cdot yr^{-1}$ when corrected for focussing (Lockhart et al. 1998). Hg analyses of the core are ongoing. If we assume a Hg concentration of 50 $ng \cdot g^{-1}$ (the average of lake sediment samples from 1998), this gives a flux rate of 4 $\mu g \cdot m^{-2} \cdot yr^{-1}$. This is a low Hg flux rate and comparable to that observed for Lac Belot and Great Bear Lake, which are low-Hg lakes. There was some evidence of slightly increased concentrations of nitrogen and inorganic and organic carbon in recent times in the core but the converse for phosphorus. These differences were small and may have been related to changes occurring within the sediments themselves rather than changing fluxes.

In summer, we focussed our initial effort on the streams flowing into Cli Lake. We located all flowing streams, measured their bank widths and depths and general water chemistry (temperature, pH, conductivity and turbidity). Streams to the west had higher conductivities than streams to the east, which were tea coloured. Streams were colder than surface lake water. A series of water samples were obtained to determine whether Hg concentrations in streams varied depending on the watershed characteristics of that stream. Concentrations in streams ranged from 1 to 3 $ng \cdot L^{-1}$ (parts per billion) while lake concentrations ranged from 0.6 to 1.0 $ng \cdot L^{-1}$. Hg concentrations in Little Doctor Lake water ranged from 1.6 to 1.9 $ng \cdot L^{-1}$ and thus were about twice as high as in Cli Lake. Concentrations appeared higher in the tea-coloured streams to the east than in streams to the west, although more samples are required for confirmation. These concentrations were in the range observed for lakes in northern Wisconsin (Watras et al. 1995) but lower than concentrations observed for lakes in the Keewatin District (Shilts and Coker 1995).

The Hg analyses of stream and lake water collected in summer 1999 have determined that Hg concentrations are higher in Cli Lake streams than the lake, and that Hg levels are higher in Little Doctor waters than Cli Lake waters. Such differences appear to be associated with water colour and, by inference, dissolved organic carbon. Dissolved organic matter plays an important role in the photoreduction of Hg (Amyot et al. 1997).

The Canadian Council of Ministers of the Environment (1999) has established a maximum acceptable concentration of 0.001 $mg{\cdot}L^{-1}$ (1 $\mu g{\cdot}L^{-1}$ or 1 part per million) in drinking water. At 1 $ng{\cdot}L^{-1}$ (part per billion), Hg concentrations in Cli and Little Doctor Lake waters are some 1000 times lower than Canadian Drinking Water Quality Guidelines.

Sediment samples were collected in several Cli Lake and Little Doctor Lake streams and analyzed by T. Jackson (National Water Research Institute, Burlington, ON) for pH, Eh (a measure of the reducing capacity of a sediment or solution) and total Hg. Most sediments were slightly acidic (pH 6.3 to 6.8) although streams to the west had more neutral (pH 7.2 to 7.8) sediments. Eh ranged from 91 to 421 mV with higher values associated with Cli Lake, particularly streams to the west. Hg concentrations in sediments ranged from 24 to 50 $ng{\cdot}g^{-1}$. Most of the Hg was tightly bound to the sediments (the magnesium chloride fraction was low) suggesting that, at these sites, very little of the Hg was available for transformation by microbes (Jackson 1989, Jackson 1991).

During the stream sampling in Cli Lake, we attempted to obtain benthic invertebrates and forage fish for Hg and stable isotope analyses. Some forage fish were obtained with minnow traps but it is possible that these fish had entered the creek mouths from the lake. Further exploration of the streams revealed a rapid narrowing a short distance upstream and increases in flow rate. It was felt that such reaches would be highly vulnerable to extensive disturbances in the search for sufficient forage fish and invertebrate biomass for Hg determinations. This aspect of the sampling was postponed pending the results of other collections from the streams.

Forage fish (nine-spined stickleback, emerald shiner, burbot, spoonhead sculpin) and benthic invertebrates (mayflies, stoneflies, leeches, snails, water boatmen, caddisflies) were collected at a number of sites in the littoral zone of Cli Lake with a smaller series of collections in Little Doctor Lake. Fish were caught by beach seine in Cli Lake. Rocks were turned over and dip nets used to collect invertebrates from rocky and weedy habitats. Periphyton and macrophytes (pondweeds, grasses) also were collected along with plankton and mysids (Cli Lake) in the offshore. Organisms in these collections have been identified, freeze-dried and lipid content determined. Stable isotope and Hg analyses are ongoing. Data will be compared with Tremblay and Lucotte (1997) and related studies.

Gill nets were set in Cli Lake and additional pike, lake trout and whitefish caught. Nets were not set in Little Doctor Lake because of time and freezer capacity considerations. Fish, including forage fish, and plankton were submitted for organochlorine, Hg and stable isotope analyses. The rate of organochlorine biomagnification will be compared with Kidd et al. (1995), Evans (1997) and later work.

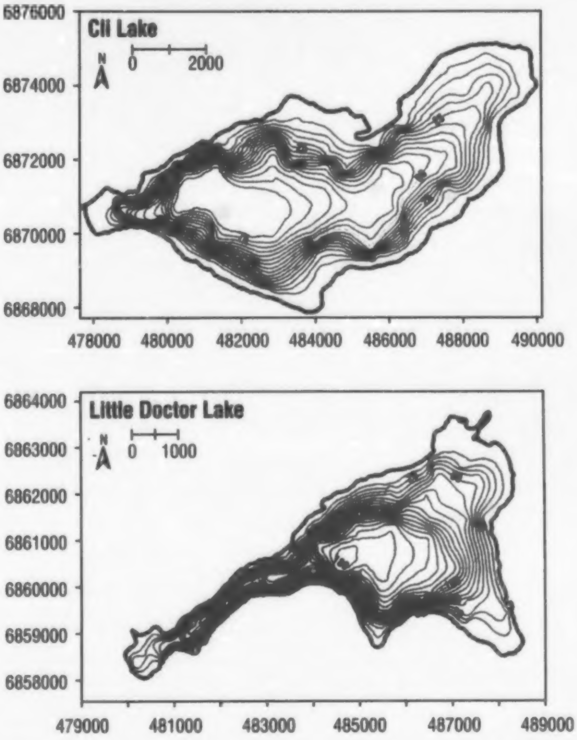
While in Little Doctor Lake, we completed our mapping of lake depths in the west end of the lake. This region was surprisingly deep with depths > 10 m. We worked with the Global Position System files from both years to develop bathymetric maps for the Cli and Little Doctor lakes. For various reasons, we were unable to conduct differential corrections. However, because we had manually recorded our locations and times, we were able, through various inferences, to develop a bathymetric map for both lakes (Figure 1).

While in Fort Simpson in July 1999, we had the opportunity to visit the Friendship Centre and to talk to students about the study. We also had a meeting with representatives of the Denendeh Resource Committee and talked with the Mayor. The Environmental Resources Committee in Jean Marie River also was visited. Both committees have requested reports summarizing the results of these studies. Summary reports are in preparation.

Conclusions

The second year of our sampling has elucidated more of the major features of Cli and Little Doctor lakes. Hg fluxes to Cli Lake sediments appear to be low. Hg concentrations in stream and lake sediments also are low. Concentrations in waters are approximately one thousand times below drinking water guidelines. Food web studies are assessing the biomagnification of total Hg in the lake ecosystem. Transformation of total Hg into biologically available forms also appears to be an essential process which requires further elucidation. While our data sets are small, it initially appears that coloured streams located on the east side of both lakes are more important in

Figure 1. Bathymetric map of Cli and Little Doctor Lakes shown in (m). Also shown are the Universal Transverse Mercator co-ordinates.



such processes than streams to the west located and flowing down the mountain slopes.

Expected Completion Date

2003

Acknowledgments

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Spatial and Long-Term Trends in Organic Contaminants and Metals in Fish Species Important to the Commercial, Sports and Domestic Fisheries of Great Slave Lake and the Slave River Ecosystem

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Abstract

This study is the first year of a long-term monitoring program of contaminant trends (PCBs, DDT, toxaphene, mercury, etc.) in lake trout, burbot (loche, maria) and pike (jack) from two regions of Great Slave Lake. The West Basin is strongly affected by inputs from the Slave River while the East Arm is influenced more by atmospheric inputs. In the West Basin, a commercial fisherman operating near the lake outflow collected the lake trout, while a Fort Resolution community member collected pike and burbot near the Slave River inflow. In the East Arm, a community member at Lutsel K'e collected the three species of fish. A commercial fisherman also provided lake trout from the Hornby Channel. Ten specimens of each species collected from the West Basin and East Arm were submitted for analyses, which are ongoing, while the remainder is archived. Presentations were given to the Great Slave Lake Advisory Board and community representatives at Lutsel K'e on previous studies conducted over

1994-1998. The Department of Fisheries and Oceans in Hay River also hosted a community presentation.

Objectives

1. To develop a cost-effective, long-term monitoring strategy for assessing temporal trends in persistent organic contaminants (including PCBs [polychlorinated biphenyls], DDT, toxaphene, coplanar PCBs) and mercury concentrations in key predatory fish species inhabiting the Great Slave Lake ecosystem, including the Slave River.
2. To assess, as part of this strategy, spatial patterns in contaminant trends and investigate possible causal factors for these patterns, including sources (long-range atmospheric transport and the Slave River) and food web pathways.
3. To integrate this monitoring program with research and monitoring programs being conducted in the Peace and Athabasca River drainage systems (supported under the Northern River Ecosystem Initiative) and G. Stern's burbot monitoring program (supported by the Northern Contaminants Program [NCP]) on the Mackenzie River at Fort Good Hope.

Introduction

The NCP noted the lack of temporal trend data for contaminants in biota as a significant knowledge gap. Accordingly, it called for the creation of a long-term biomonitoring program to establish time trends in contaminant concentrations. Furthermore, the NCP identified fish as the best organism for the biomonitoring of contaminant trends in freshwater environments. A number of freshwater sites were proposed and then incorporated into this biomonitoring program, including Great Slave Lake, the focus of this study. Biomonitoring in the Slave River will be conducted in 2000-2001.

There were several reasons why Great Slave Lake, including the Slave River, was selected as an essential feature of this biomonitoring program. In brief, they are outlined below.

First, detailed contaminant biomagnification studies were conducted on Great Slave Lake during 1993-1996 (Evans 1994, Evans 1996, Evans et al. 1996, Evans et al. 1998). There also was the intensive Slave River Monitoring Program that extended from 1991 to 1994 (Sanderson et al. 1998) and other related studies (Jackson et al. 1996, LaFontaine 1997, McCarthy et al. 1997). Fish samples and extracts are archived from the Evans et al. and Sanderson et al. studies. Archived samples can be analyzed for compounds that were not investigated in the original study or to compare differences between current analytical methods and older methods. Archived samples are highly useful for long-term monitoring studies and provide for a greater confidence in utilizing older data (Huestis et al. 1996).

Second, Great Slave Lake is a major population center in the Northwest Territories. Yellowknife is the territorial capital, Hay River is an important transportation hub, and there are numerous other communities along the lake including Fort Resolution, Lutsel K'e, and Rae (North Arm). Fish populations are very important to the region. Furthermore, there has been a commercial fishery on Great Slave Lake since the early 1950s.

Third, while long-term records have been maintained on the commercial fisheries, few studies have investigated contaminant concentrations in the fisheries. In contrast, there are extensive long-term data sets for contaminant trends in Great Lakes fish (Hesselberg et al. 1990, Suns et al. 1993, Huestis et al. 1996). It is timely that similar such data sets be generated for important Great Slave Lake fish.

Fourth, Great Slave Lake is strongly influenced by the Slave River that is formed by the confluence of the Peace and Athabasca rivers. Increased development in the Peace Athabasca drainage basins (oil sands, pulp and paper mills, agriculture, urban areas, etc.) potentially may cause an increase in contaminant loading to Great Slave Lake.

Lastly, in contrast to most aquatic systems in the Northwest Territories, Great Slave Lake is well studied; the major features of its limnology and fisheries ecology are well understood. Classic limnological and fisheries studies were conducted by Rawson (1950, 1951, 1953, 1956), and much limnological data, including nutrients, stable isotopes and plankton community structure has been obtained by M. Evans during the course of her sampling.

Activities in 1999-2000

The Great Slave Lake long-term biomonitoring program has an excellent beginning. In Fort Resolution, in the West Basin and near the Slave River outflow, Gab Lafferty collected 20 pike (jack) and 11 burbot in summer as part of a fish health study. He then collected 20 burbot (loche, maria) in winter. These summer and winter collections of burbot will provide for a seasonal comparison of contaminant levels, stable isotope composition, etc. in burbot. All fish were shipped frozen to Saskatoon where they were catalogued, weighed and approximate lengths were determined.

This project calls for the analysis of 10 specimens of each species from the West Basin and the East Arm.

Therefore, 10 pike and five summer-caught and five winter-caught burbot from Fort Resolution were selected for analyses and the remainder archived. A representative sample of each collection was selected based on fish weight and approximate length.

Lake trout are infrequent in the shallow, nearshore waters of the West Basin during summer (burbot are also less common than in winter). In late autumn, Don Sinclair (Hay River) provided 20 lake trout caught near Slave Point during his commercial fishing. We also obtained 10 lake trout from the Hornby Channel to the east of the Slave River delta at about the same time. We are uncertain whether trout collected in this region are residents of the East Arm and moved into the area, residents of the West Basin and moved into the channel, or are resident to the channel. Therefore, we selected 10 of the 20 lake trout from Slave Point and archived the remainder along with the Hornby Channel lake trout.

In the East Arm, Ernest Boucher collected 20 lake trout, 20 pike and 25 burbot from offshore of Lutsel K'e during summer. We selected 10 specimens of each species for analyses and archived the remainder.

Intact fish were shipped to the Department of Fisheries and Oceans, Burlington, for initial examination (final length determinations, sex), tissue preparation and extraction. Shipment occurred after the entire collection series had been completed. Burbot liver and lake trout and pike muscle are being analyzed for persistent organic pollutants including PCBs, DDT, dieldrin and toxaphene. A subset also is being examined for coplanar PCBs and being prepared for possible future dioxin/furan analyses. Muscle is also being analyzed for metals (mercury, arsenic, and selenium in muscle) and carbon and nitrogen isotope analyses. The latter analyses will provide information on carbon sources and trophic feeding. Chemical analyses are ongoing.

Following the removal of the appropriate tissues from the individual fish, the remaining fish was homogenated; homegenates were placed in several 500-mL containers for archiving. These samples were shipped back to the National Water Research Institute in Saskatoon. Because relatively small amounts of tissue were removed from most fish, these homogenates represent "whole body." The smaller burbot from Lutsel K'e is an exception because liver is a large fraction of burbot body weight and all the liver had to be used for smaller fish. At some later date, we could compare contaminant determinations on specific tissue samples (of interest from a consumption viewpoint) and whole body (of interest in the biomagnification of contaminants from one organism to another).

Results and Discussion

Fish collected from different regions of Great Slave Lake were similar in length and weight (Table 1). Pike from Lutsel K'e and Resolution Bay were particularly similar in mean weight and length. Winter-caught lake trout from Slave Point were slightly smaller, on average, than trout from Lutsel K'e and the Hornby Channel. Burbot caught from Resolution Bay were larger than burbot caught from Lutsel K'e. Burbot are uncommon in the East Arm and larger specimens tend to be found near major river mouths such as the Slave and Yellowknife River (Rawson 1951). Resolution Bay burbot tended to be larger in winter than summer.

Table 1. Summary statistics for fish caught in Great Slave Lake, 1999-2000

	Location	Season	Number	Length (mm)	Weight (g)
East Arm					
Lake trout	Lutsel K'e	Summer	20	609 ± 69	3096 ± 1239
Pike	Lutsel K'e	Summer	20	660 ± 158	2348 ± 1403
Burbot	Lutsel K'e	Summer	25	480 ± 68	884 ± 420
West Basin					
Lake Trout	Hornby Channel	Autumn	10	621 ± 46	3094 ± 788
Lake Trout	Slave Point	Winter	20	578 ± 36	2528 ± 486
Pike	Resolution Bay	Summer	22	670 ± 111	2576 ± 1303
Burbot	Resolution Bay	Summer	11	628 ± 58	1637 ± 436
Burbot	Resolution Bay	Winter	20	678 ± 48	2250 ± 556

Table 2. Summary statistics for lipid and moisture content of muscle and the gender of fish from Table 1 which are being analyzed for organic contaminants and metals

	Location	Time	Number	% moisture	% Lipid	% female
East Arm						
Lake trout	Lutsel K'e	Summer	10	73.7 ± 4.0	5.2 ± 3.5	20
Pike	Lutsel K'e	Summer	10	79.5 ± 1.0	1.0 ± 0.7	70
Burbot	Lutsel K'e	Summer	10	82.4 ± 1.2	2.1 ± 1.3	10
West Basin						
Lake Trout	Slave Point	Winter	10	73.3 ± 1.8	7.9 ± 1.6	10
Pike	Resolution Bay	Summer	10	79.3 ± 1.0	0.9 ± 0.3	60
Burbot	Resolution Bay	Summer	5	81.4 ± 1.5	3.7 ± 0.9	60
Burbot	Resolution Bay	Winter	5	79.3 ± 4.2	5.6 ± 3.5	80

Males dominated the lake trout catch from both locations (Table 2). Lake trout had the highest lipid content of the three species examined. Winter-caught West Basin fish were slightly more lipid-rich than summer-caught females from the East Arm. Summer-caught pike were similar in lipid content from both locations. There were slightly more males than females in the catch. Burbot from the West Basin had a slightly higher lipid content than burbot from the East Arm; lipid content was greater in winter. Females dominated the West Basin catches and males the East Arm.

There were a number of formal and informal communications. Informal communications were based on telephone calls regarding sample collection and general questions. Collections at Lutsel K'e and Fort Resolution were, in part, coordinated with other fisheries projects in the communities. George Low was integrally involved with these studies and also worked with the commercial fishermen.

George Low also facilitated a presentation by M. Evans to the Great Slave Lake Advisory Board on earlier and ongoing Great Slave Lake studies. The Board is comprised of commercial fishermen, community representatives and government agency representatives. This presentation was given in December 1999 in Yellowknife. Following the presentation, M. Evans was interviewed by Rose Marie Blackduck of CBC Radio and two days later by Randy Henderson, also with CBC Radio. During both broadcasts, special note was made at the beginning of the fact that contaminant levels tend to be higher in organisms in the East Arm than in the West Basin. Lutsel K'e requested a community visit following this broadcast to explain this information more fully. In March, M. Evans visited the community and discussed the results of the previous and ongoing Great Slave Lake studies to a number of community representatives. The Department of Fisheries and Oceans, Hay River, helped organize and host a similar presentation in Hay River.

During the year, data generated from the original Great Slave Lake study have been compiled into larger data files for final synthesis. Because very large data sets are involved, this stage invariably involves some re-checking. The fish data will be placed into a brief report that will summarize these earlier data and the contaminant data from the 1999-2000 studies. In addition, comparisons will be provided with contaminant levels in similar fish from the Laurentian Great Lakes. By providing brief reports, we hope to streamline the rate at which contaminant data become available to the communities and for health assessments.

Conclusions

The first year of this study has had a good start. Three species of fish were successfully collected from two major regions of Great Slave Lake. It was hoped that collection efforts could be completed in summer 1999 in order to integrate this study with other ongoing fisheries studies in the communities. This also could allow for the chemical analyses to be completed during winter 2000. However, the West Basin is shallow and fish such as lake trout and burbot are difficult to collect in large numbers as part of ongoing fisheries. Adjustments in the collection effort are being considered.

Preliminary data suggests that the fish collected in this study were relatively similar to one another between the two regions of the lake and summer versus winter. The notable exception was the burbot from Lutsel K'e which were smaller than from Resolution Bay.

The results of this study and earlier studies are of interest to the communities and communications are good. The publication of a brief report summarizing the major features of the fish data will improve these communications and help streamline the consumption assessments.

Expected Completion Date

This is on ongoing monitoring study. Annual sampling is projected until winter 2002 and analysis of collected fish until 2003. It is further anticipated that monitoring will continue after NCP Phase II is completed.

Acknowledgments

The contributions of the Project Team are gratefully acknowledged and have been noted in this report. Don Sinclair provided the lake trout from Slave Point and Steve Blakely the lake trout from the Hornby Channel commercial fishery. Fred Taptuna, with Fisheries and Oceans, Hay River arranged and hosted the March community presentation. Lucy Sanderson, Lutsel K'e, hosted the March community presentation in Lutsel K'e. Special appreciation is extended to Gab Lafferty and Ernest Boucher for collecting the fish from Lutsel K'e and Resolution Bay. Their close attention to detail and dedication in obtaining these fish is very much appreciated.

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Mercury in Fish from Stock Surveys of Lakes in the Western Northwest Territories: Investigations into the Factors Affecting Mercury Levels

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Abstract

Surveys of limited numbers of fish from lakes in the Northwest Territories and Nunavut since the 1970s have indicated that fish from some lakes contain fish with levels of mercury above those recommended for human consumption. This raises questions about the geographic extent of the problem of high mercury levels, the sources of the mercury, whether the problem is entirely natural or whether there is an anthropogenic component, and whether the magnitude of the problem is changing. For several years, fish taken in Northern Contaminants Program projects have been analyzed for mercury. More recently, fish taken in stock surveys carried out at the request of Renewable Resource Councils have been the source of fish for this project. The result is a growing database of mercury in fish from lakes chosen either by other Northern Contaminants Program projects or by local communities. The data has revealed that different species generally contain different levels of mercury, with predators such as lake trout, walleye and northern pike usually containing more mercury than forage feeders such as cisco and whitefish. Some of the levels in predator species of fish have been found to exceed levels considered safe for unrestricted human consumption and consequently some consumption advisories have been issued. The data from two of the lakes reported this year may prompt the issuance of more advisories. The reasons why levels of a given species are higher in one lake than

another are not yet clear and are the subject of ongoing investigations. Comparison of levels in fish with a series of regional geological maps suggests that geological data at a much finer scale will be necessary to understand the influence of regional geology on mercury levels in fish from different lakes.

Objectives

1. To determine the occurrence and geographic distribution of mercury (Hg), selenium (Se) and arsenic (As) in fish from those inland lakes in the Northwest Territories being assessed by the Department of Fisheries and Oceans (DFO) for their ability to produce sustained yields of fish for subsistence consumption by Northern people.
2. To evaluate the levels of Hg found in the fish in terms of the age/size relationships of the fish and also in terms of the geological setting of each lake and its drainage, with a view to developing a management strategy for recommending levels of consumption based on species and size of fish on a lake-by-lake basis.

Introduction

Under the terms of land claims settlements, DFO is responsible for the conduct of studies on fish populations at the request of the Renewable Resource Councils in order to assess the existing or proposed harvest of stocks for subsistence, commercial or tourist uses. Historical DFO Inspection Service data indicate that muscle Hg levels in some of the larger fish from a number of lakes in the Northwest Territories exceed levels deemed safe by Health Canada for human consumption. However, the sample sizes are small and samples were generally biased to size classes typical of commercial catches.

Fish from a number of lakes have been surveyed over the past several years. The levels of Hg in the fish differ from lake to lake and conclusions drawn from one lake do not necessarily apply to another lake. Fish that feed near the base of the food chain (whitefish, cisco, sucker) consistently contain less Hg those that feed near the top of the food chain (walleye, pike, lake trout). In a number of instances, the predators contained levels high enough to require consumption advisories (Lockhart et al. 1999a, Lockhart et al. 1999b). We have begun to investigate the form of Hg found in the fish and results to date indicate that most of it is methylmercury (MeHg).

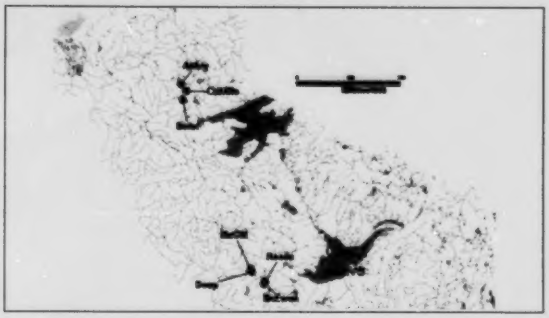
Several factors influence the level of Hg in fish tissues, including fish age (or length), the structure of the supporting food chain, water chemistry, lake temperature, microbiological processes in the sediments, fish biochemistry, the supply of Hg to the lake and the nature of the drainage (e.g. rocks, uplands, wetlands, etc.). The supply of Hg is a mixture of natural geological contributions and human-related contributions. There is no doubt that gaseous Hg moves with air masses because concentrations in air have been measured at Alert, Nunavut (Schroeder et al. 1998) and further that Hg is found in snow falling in the Arctic (Lu et al. 2000). These studies, however, do not answer the question about the relative importance of natural and anthropogenic sources of Hg. To a surprising extent, the answer to this question remains unknown, although lake sediment core studies suggest that in some locations, anthropogenic inputs may be as large as natural inputs. Unfortunately, Hg does not have isotopic mixtures convenient for discriminating among different sources. When survey data from northern lakes are combined with sediment core studies, geological settings, limnological information and experimental studies now underway at the Experimental Lakes Area, we should be able to estimate the amounts of Hg derived from the various sources.

Several independent lines of evidence suggest that inputs of Hg to northern Canada have increased over pre-industrial levels. Hg in Arctic seals and whales has increased since the 1970s (Wagemann et al. 1996). Sediment core data from a number of lakes in the Northwest Territories have suggested that recent Hg inputs are substantially above pre-industrial levels (Lockhart et al. 1998). Recently Braune (1999) reported that Hg levels have also increased in seabirds. Given these considerations, it is important to assess the levels of Hg in stocks of fish being used now or being assessed for potential harvest in the near future.

Activities in 1999-2000

Collections were made during 1999-2000 to supplement the data on hand from previous studies. The lakes in the Northwest Territories for which new samples were collected were: McEwan, Reade, McGill, Deep, Colville, Aubry and Belot. A map showing the locations of these lakes is shown in Figure 1. Two of these (Colville and Belot) were sampled previously in 1993. Some additional collections made early in 2000 are still being analyzed. We have concentrated on the analyses for Hg with the result that some of the As and Se analyses are still being done. All concentrations in fish tissues are expressed as weight of contaminant per gram of wet tissue weight. Digital geological maps

Figure 1. Map showing the locations of the seven lakes from which fish have been obtained for this project



of the area have been examined for correlations with Hg measurements.

Results and Discussion

McEwan Lake (60° 49' N, 119° 57' W)

One hundred and six fish were obtained from McEwan Lake in January 2000. These comprised seven burbot, 36 cisco, 30 lake whitefish, 15 northern pike and 18 walleye. Mean Hg levels were as follows:

Species	Mean muscle Hg concentration ± standard deviation
Burbot	0.086 ± 0.033 (n = 7)
Cisco	0.093 ± 0.028 (n = 35, 1 missing)
Lake whitefish	0.088 ± 0.035 (n = 30)
Northern pike	0.331 ± 0.238 (n = 15)
Walleye	0.356 ± 0.158 (n = 17, 1 missing)

As expected, the predators (northern pike and walleye), had strikingly higher levels than the forage feeders (lake whitefish and cisco). In this instance, burbot were similar to the forage feeders although in some instances they are more like other predators.

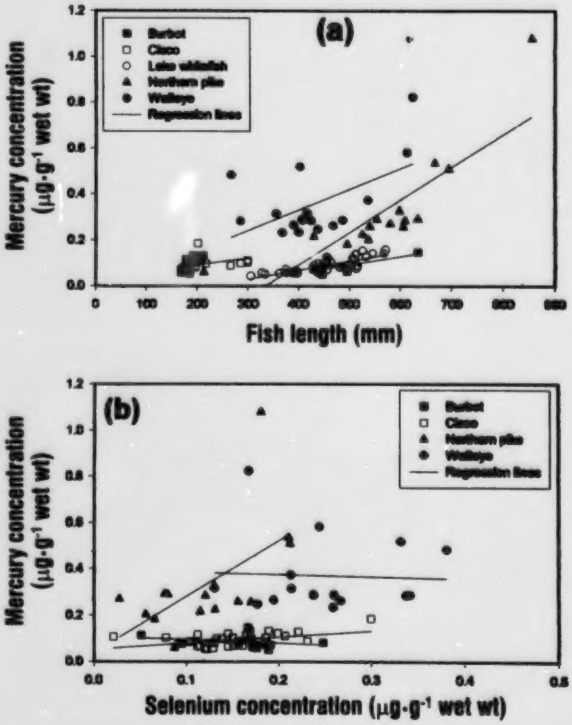
Graphs showing the levels of Hg in fish of differing length are shown in Figure 2a. Generally, the larger fish had higher Hg concentrations. The tendency for Hg concentration to increase with length was described using linear regression equations calculated with NCSS-2000 statistical software. The regression coefficient was statistically significant ($p < 0.05$) for all species but burbot. The r^2 value was high for burbot, but there were only seven samples. The regression equations described most of the variation in the data for lake whitefish and northern pike.

Species	Regression equation	r^2
Burbot	Hg ($\mu g \cdot g^{-1}$) = 0.000345 × Length (mm) – 0.0794	0.565
Cisco	Hg ($\mu g \cdot g^{-1}$) = 0.000264 × Length (mm) + 0.0402	0.112*
Lake whitefish	Hg ($\mu g \cdot g^{-1}$) = 0.000397 × Length (mm) – 0.0907	0.710*
Northern pike	Hg ($\mu g \cdot g^{-1}$) = 0.001471 × Length (mm) – 0.4748	0.689*
Walleye	Hg ($\mu g \cdot g^{-1}$) = 0.000896 × Length (mm) – 0.0276	0.299*

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

Se was measured in most of the same samples and the relationships between Hg and Se are shown graphically in Figure 2b. There was no statistical relationship between these elements in burbot, but there was a significant tendency for increasing Hg with increasing Se in cisco ($r^2 = 0.208$*). Hg and Se levels in northern pike were related ($r^2 = 0.331$*) but not in walleye. Analyses for As for these fish are incomplete.

Figure 2. Mercury in fish from McEwan Lake, NWT, January 2000*



* The top panel (a) describes muscle mercury concentration in terms of fish length. The bottom panel (b) shows the same mercury data plotted as a function of muscle selenium. Selenium results for lake whitefish are not available yet.

Reade (unnamed) Lake (60° 54' N, 119° 55' W)

Reade Lake was sampled in January 2000, and 77 fish were taken: two burbot, 30 lake whitefish and 45 northern pike. The mean level of Hg in muscle of the two burbot was 0.086 $\mu g \cdot g^{-1}$ while that for the 30 lake whitefish was 0.146 ± 0.044 $\mu g \cdot g^{-1}$ and for northern pike, 0.430 ± 0.203 $\mu g \cdot g^{-1}$. Larger whitefish and northern pike had higher Hg levels than smaller ones (Figure 3a). The equations relating size and Hg concentration are shown below.

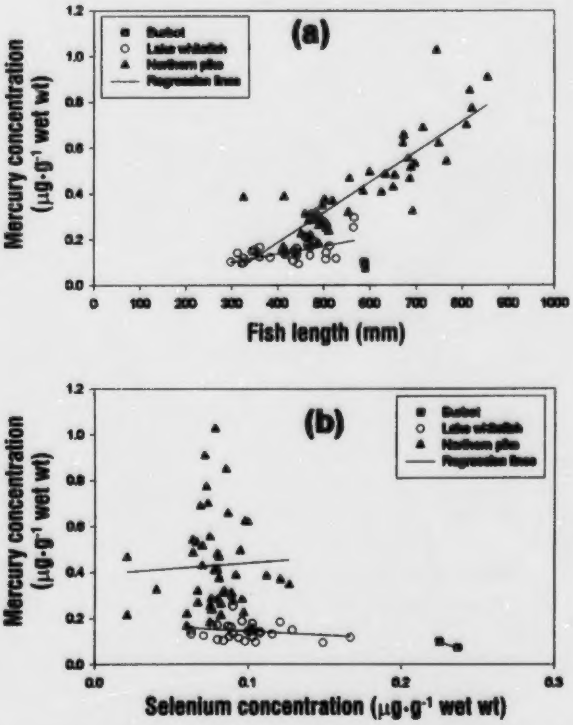
Species	Regression equation	r^2
Lake whitefish	Hg ($\mu g \cdot g^{-1}$) = 0.000342 × Length (mm) – 0.000469	0.346*
Northern pike	Hg ($\mu g \cdot g^{-1}$) = 0.001326 × Length (mm) – 0.3434	0.720*

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

There were no significant statistical associations between Hg and Se (Figure 3b).

As levels were determined in the Reade Lake fish and they were uniformly low. All but two of the northern pike had levels below our limit of quantitation of 0.05 $\mu g \cdot g^{-1}$. The lake whitefish had levels just above detection, in the range of 0.07 $\mu g \cdot g^{-1}$while the two burbot had muscle As levels of 0.05 and 0.055 $\mu g \cdot g^{-1}$.

Figure 3. Mercury in fish from Reade (unnamed) Lake, NWT, January 2000*



* The top panel (a) describes muscle mercury concentration in terms of fish length. The bottom panel (b) shows the same mercury data plotted as a function of muscle selenium.

McGill Lake (61° 18' N, 121° 0.75' W)

McGill Lake was sampled in February 2000, and 97 fish were obtained for this project: one burbot, 29 lake whitefish, 28 northern pike, 22 walleye and 17 white suckers. The mean concentrations of Hg in muscle of these fish are shown below.

Species	Mean muscle Hg concentration ± standard deviation
Burbot	0.104 (n=1)
Lake whitefish	0.150 ± 0.068 (n=29)
Northern pike	0.713 ± 0.365 (n=28)
Walleye	1.125 ± 0.378 (n=22)
White sucker	0.206 ± 0.081 (n=17)

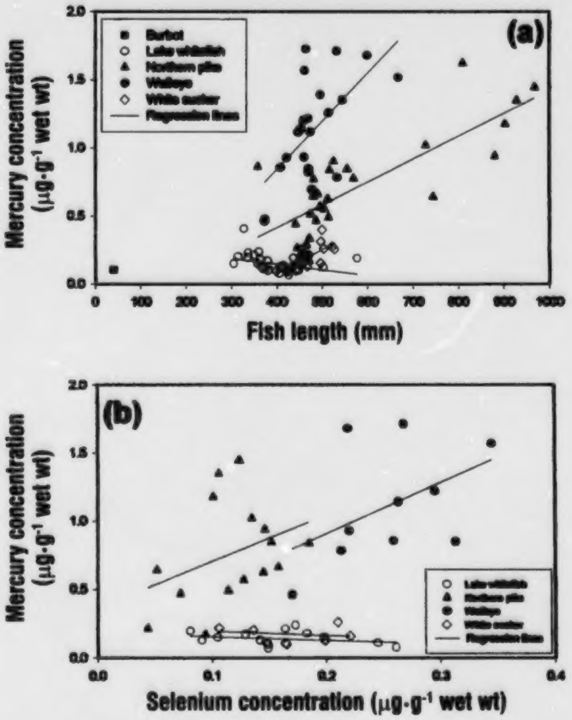
Mean values of Hg for both northern pike and walleye exceeded the guideline level of 0.5 $\mu g \cdot g^{-1}$ used to regulate commercial sale of fish in Canada. Concentrations of Hg in muscle are shown graphically in Figure 4a for all the fish as related to fish length. Generally the levels of Hg increased with increasing length, with the exception of lake whitefish. The regression equations incorporating Hg and length are listed below.

Species	Regression equation	r^2
Burbot	not applicable, n = 1	
Lake whitefish	Hg ($\mu g \cdot g^{-1}$) = -0.00041 × Length (mm) + 0.3103	0.119
Northern pike	Hg ($\mu g \cdot g^{-1}$) = 0.00168 × Length (mm) – 0.2543	0.617*
Walleye	Hg ($\mu g \cdot g^{-1}$) = 0.00353 × Length (mm) – 0.5701	0.396*
White sucker	Hg ($\mu g \cdot g^{-1}$) = 0.00173 × Length (mm) – 0.6102	0.512*

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

Se and As analyses of these fish are still underway. The Se values available to date are shown together with the Hg values for the same fish in Figure 4b. There were no significant statistical relationships between Hg and Se for any species, although such relationships may still be identified when the remaining Se analyses are available. Most of the As concentrations found were below 0.05 $\mu g \cdot g^{-1}$; of the few that were above detection, the highest was 0.081 $\mu g \cdot g^{-1}$.

Figure 4. Mercury in fish from McGill Lake, NWT, February 2000*



* The top panel (a) describes muscle mercury concentration in terms of fish length. The bottom panel (b) shows the same mercury data plotted as a function of muscle selenium.

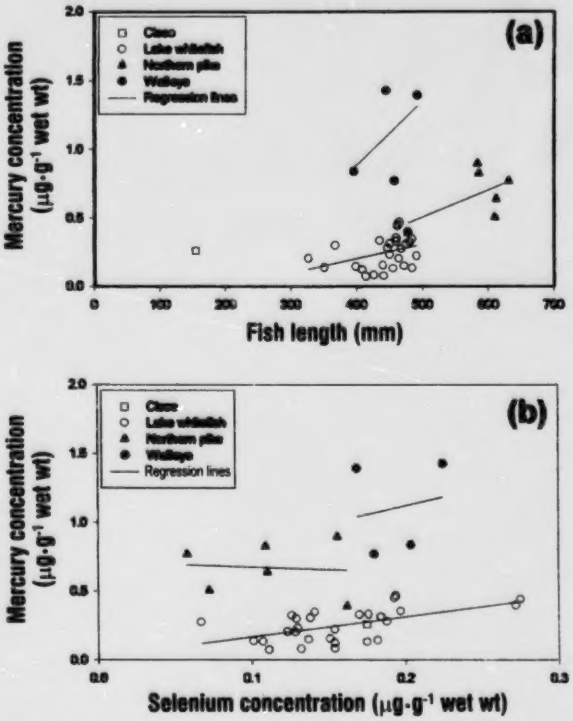
Deep Lake (61° 12.5' N, 120° 54.5' W)

Thirty-nine fish were obtained from Deep Lake in February 2000: one cisco, 28 lake whitefish, six northern pike and four walleye. Mean Hg concentrations in muscle were:

Species	Mean muscle Hg concentration ± standard deviation
Cisco	0.260 (n=1)
Lake whitefish	0.249 ± 0.119 (n=28)
Northern pike	0.670 ± 0.196 (n=6)
Walleye	1.105 ± 0.352 (n=4)

The numbers of northern pike and walleye were quite small but the mean Hg levels for these species were both quite high (over the recommended maximum for commercial sale of fish). The Hg values are plotted against fish length in Figure 5a and, while there appear to be higher levels of Hg in larger fish, none of the slopes was statistically different from zero.

Figure 5. Mercury in fish from Deep Lake, NWT, February 2000*



* The top panel (a) describes muscle mercury concentration in terms of fish length. The bottom panel (b) shows the same mercury data plotted as a function of muscle selenium.

The relationships between Hg and Se are shown in Figure 5b. Lake whitefish with higher Se levels did have slightly higher Hg levels ($r^2 = 0.321$, $p < 0.05$). There were too few northern pike and walleye to support conclusions about relationships between these two elements.

Arsenic concentrations have been determined in muscle of all these fish and they were low. Most of the results were below detection with the maximum concentration found being 0.144 $\mu g \cdot g^{-1}$.

Colville Lake (67° 19' N, 126° 0' W)

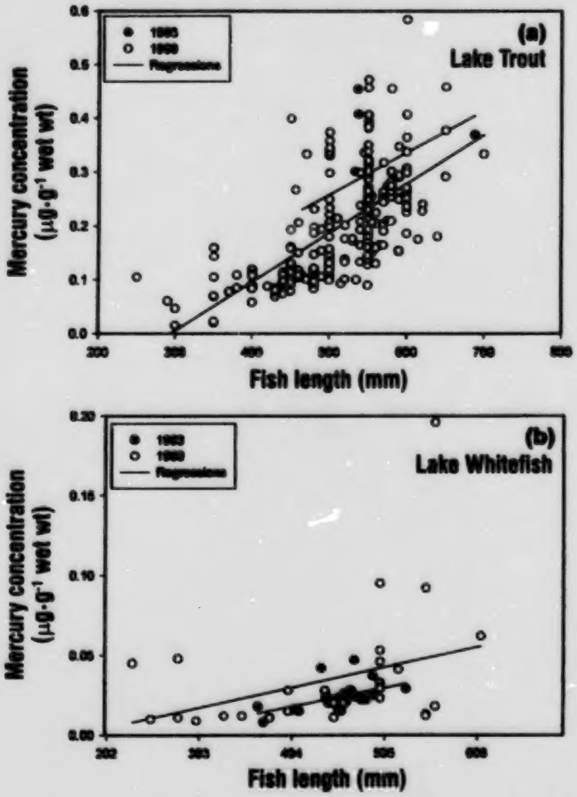
We obtained some samples of fish from Colville Lake in 1993 and again in 1999. The numbers and species are shown below:

Species	Number in 1993	Number in 1999
Burbot	0	18
Lake trout	10	240
Lake whitefish	24	30
Northern pike	0	7

Although the number of samples in 1993 was small, these collections offer the possibility to comment on temporal change. The average concentration of Hg in 1999 was calculated from 238 fish because length data were not obtained from two of them. The mean concentration of Hg in muscle of the 10 lake trout in 1993 was 0.307 ± 0.103 $\mu g \cdot g^{-1}$ while the average for the 238 fish in 1999 was only 0.199 ± 0.103 $\mu g \cdot g^{-1}$. Figure 6a shows the graph of muscle Hg as a function of length for both samples and it is evident that larger fish tended to contain more Hg. There were consistently higher levels of Hg in the larger fish in 1999 but this relationship was equivocal in the smaller sample in 1993 ($r^2 = 0.192$, $p = 0.205$). The slopes of regression lines through the two sets of data are included in Figure 6a and these appear to be similar. The equation describing Hg in terms of length for the fish in 1999 was:

Species	Regression equation	r^2
Lake trout (1999)	Hg ($\mu g \cdot g^{-1}$) = 0.000909 × Length (mm) – 0.2673	0.42

Figure 6. Concentrations of mercury in muscle of lake trout and whitefish from Colville Lake*



* The top panel shows concentrations in lake trout in 1993 (n = 10) and in 1999 (n = 238). The bottom panel shows concentrations in lake whitefish in 1993 (n = 24) and 1999 (n = 30).

The fish taken in 1993 were considerably larger than the recent sample; the mean length for the 10 fish in 1993 was 562.4 mm as compared with 511.9 mm in 1999. Given the difference in size of the fish between 1993 and 1999, an analysis of covariance was carried out to test whether the levels of Hg had changed. This analysis failed to detect a difference in slopes for the two sets of data plotted in Figure 6a and indicated that the adjusted mean for 1999 was in fact somewhat lower than that in 1993 (F = 6.11, df = 1243). This may indicate that the levels of Hg in Colville Lake have declined slightly or it may be an artifact of the small sample size in 1993. A decline in levels would be inconsistent with other Arctic data on whales and sea birds (Wagemann et al. 1996, Braune 1999). Two sediment cores were obtained from Colville Lake in 1993 and, while they cannot explain differences between 1993 and 1999, they both showed increases in Hg in the most recent slices relative to conditions in the early years of the 20th century. Core 1 showed a small increase in the early 1990s relative to the 1980s but core 2 showed a small decrease in the same period (Lockhart et al. 2000, this volume). There have been indications of recent declines in atmospheric deposition of Hg in some areas (Engstrom and Swain 1997) but we have not seen that indication yet in data from the Canadian Arctic. The database contains a few instances in which the same lake has been sampled on more than one occasion. For example, five lake trout from Cli Lake were sampled in 1983 and 49 were sampled in 1996. The sizes of the fish were similar on both occasions (mean length was 477 mm in 1983 and 485 mm in 1996). The mean Hg level in 1983 was 0.39 $\mu g \cdot g^{-1}$ and it was 0.88 $\mu g \cdot g^{-1}$ in 1996.

Lake whitefish were obtained in 1993 and again in 1999 and so this species also offered and opportunity to examine changes during the 6-year interval. Whitefish had lower concentrations of Hg in muscle than the other species sampled. The mean Hg level in 1993 was 0.024 ± 0.0087 $\mu g \cdot g^{-1}$ (n = 24) while in 1999 it was 0.035 ± 0.0378 $\mu g \cdot g^{-1}$ (n = 30). The data are plotted in Figure 6b and the slopes for the two collections were similar (F = 0.004, $p > 0.05$). The equations describing the relationships between length and Hg levels are:

Species	Regression equation	r^2
Lake whitefish (1993)	Hg ($\mu g \cdot g^{-1}$) = 0.000115 × Length (mm) – 0.0286	0.235*
Lake whitefish (1999)	Hg ($\mu g \cdot g^{-1}$) = 0.000125 × Length (mm) – 0.0206	0.119

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

The slope for the collection in 1999 fell just short of the statistical criterion of $p < 0.05$, the probability being 0.06. Given the similarity of the slopes, the means were compared by analysis of covariance and they failed to differ (F = 2.8, $p = 0.10$).

We also obtained 18 burbot and seven northern pike in 1999. The average concentration of Hg in muscle of burbot was 0.116 ± 0.041 $\mu g \cdot g^{-1}$ while that in northern pike was 0.244 ± 0.035 $\mu g \cdot g^{-1}$. The burbot displayed a mild tendency towards higher Hg concentrations in larger fish:

Species	Regression equation	r²
Burbot	Hg ($\mu g \cdot g^{-1}$) = 0.0002458 × Length (mm) – 0.0301	0.267

With northern pike, however, the sample number was very small and there was no clear trend of co-variation of Hg and size. As and Se values are not available yet for fish from Colville Lake.

Aubry Lake (67° 24' N, 126° 27' W)

The collection of fish from Aubry lake in November/December 1999 yielded 96 lake trout and eight burbot. Hg analyses were completed for 88 of the lake trout and all burbot. The average muscle Hg level in lake trout was 0.254 $\mu g \cdot g^{-1}$ and the relationship between length and Hg is shown in Figure 7. The equation describing the relationship between length and Hg concentration is:

Species	Regression equation	r²
Lake trout	Hg ($\mu g \cdot g^{-1}$) = 0.0008605 × Length (mm) – 0.2428	0.49*

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

The average muscle concentration in the burbot was much lower than the lake trout, 0.108 ± 0.0196 $\mu g \cdot g^{-1}$, and the relationship between muscle Hg and length in burbot was not significant. As and Se analyses of these fish are incomplete.

Lac Belot (66° 53' N, 126° 16' W)

Lake trout from Lac Belot were sampled in April 1993, and again in December 1999. The mean muscle Hg level found in 1993 was 0.135 ± 0.049 $\mu g \cdot g^{-1}$ (n = 19) as compared with 0.212 ± 0.075 $\mu g \cdot g^{-1}$ (n = 54) in 1999. The fish samples were concentrated in a relatively narrow range of lengths, with average lengths on the two occasions similar at 621.3 mm in 1993 and 617.6 mm in 1999. The Hg concentrations are shown in Figure 8 and the least squares regression lines described by the equations below are included on the figure.

Species	Regression equation	r²
Lake trout (1993)	Hg ($\mu g \cdot g^{-1}$) = 0.0004473 × Length (mm) – 0.1435	0.123
Lake trout (1999)	Hg ($\mu g \cdot g^{-1}$) = 0.0007005 × Length (mm) – 0.2204	0.274*

* Indicates statistical significance of the regression coefficient at $p < 0.05$ or less.

The slope for the samples in 1999 was statistically significant but that for 1993 was not. Analysis of covariance indicated that the means for the 2 years were significantly different (F = 24.52, $p < 0.001$), implying an increase in Hg levels between 1993 and 1999. Given the close proximity of Lac Belot to Colville Lake, it seems unlikely that the Hg in the two lakes would show different trends if the trends are driven either by local geological activities or by atmospheric fallout.

Six burbot were also obtained in 1999 and these had a mean muscle Hg content of 0.130 ± 0.039 $\mu g \cdot g^{-1}$.

Figure 7. Mercury in muscle of fish from Aubry Lake, NWT, November-December 2000, as functions of fish length

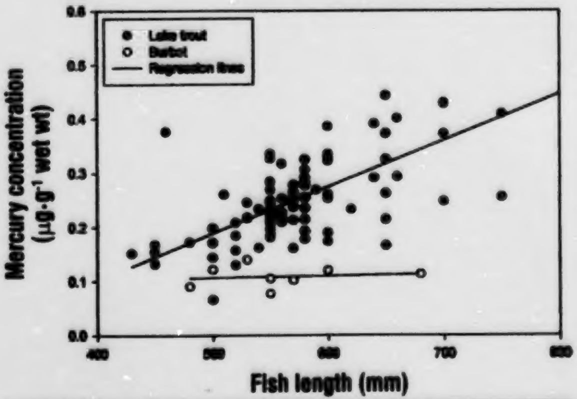
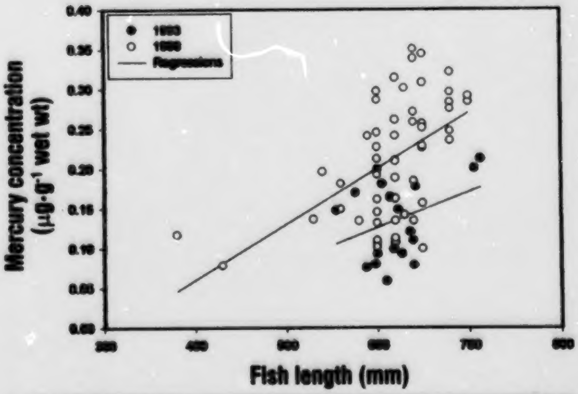


Figure 8. Concentrations of mercury in muscle of lake trout from Lac Belot in 1993 (n = 19) and in 1999 (n = 54)



Comparison of the mercury levels in fish from the seven lakes

The mean concentrations of Hg in muscle of fish from the seven lakes are shown graphically in Figure 9. The most striking concentrations are those from McGill and Deep Lakes for northern pike and walleye. Rigorous comparisons are difficult because the mix of species differs among the lakes and the means as shown are not adjusted for fish of differing sizes. We have whitefish and burbot from five of the seven lakes. The four small southern lakes all had whitefish with higher Hg levels than those from Colville Lake. However, Hg in burbot was relatively similar across all the lakes.

We have a growing database on Hg in fish and Figure 10 shows a map describing levels in lake trout from a wide geographic range of lakes. The range of mean values represented by these fish was from 0.08 $\mu g{\cdot}g^{-1}$ (Lake Tunago, n = 1, 1978) to 1.04 $\mu g{\cdot}g^{-1}$ (Ferguson Lake, n = 10, 1971) (excluding sites influenced by the Discovery mine). Hence there is about a 10-fold range in Hg levels in fish from sites with no known local sources. Comparing this range of means with that for polychlorinated biphenyls (PCBs), the *Canadian Arctic Contaminants Assessment Report* (Jensen et al. 1997) tabulated a range of means for PCBs in lake trout muscle from 8.79 to 448 $ng{\cdot}g^{-1}$, a range of about 50-fold. If Lake Laberge is dropped from the PCB comparison, the range falls to about five-fold. From this comparison, it is evident that the range of variation of Hg levels in the fish is comparable to that for PCBs, which have no natural sources and for which the main pathway to northern lakes is via the atmosphere. It is possible then that a substantial share of the variation in lake-to-lake Hg levels may be attributable to atmospheric sources.

Figure 9. Comparison of mean concentrations of mercury in fish from the seven lakes sampled during 1999-2000

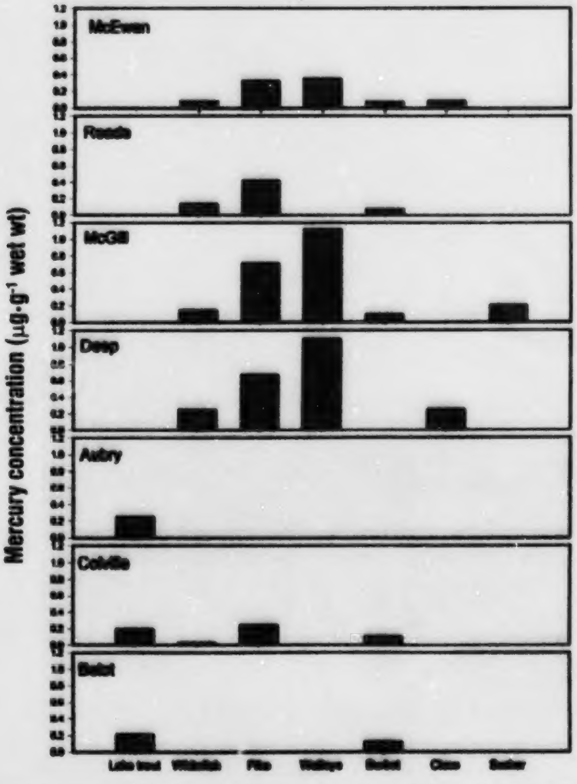
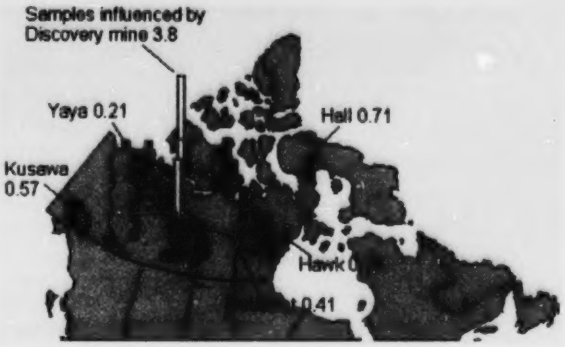


Figure 10. Mean concentrations of mercury ($\mu g{\cdot}g^{-1}$)in muscle of lake trout from northern Canadian lakes*



* Source: The former DFO Inspection Service, this project and several other independent studies by a number of investigators

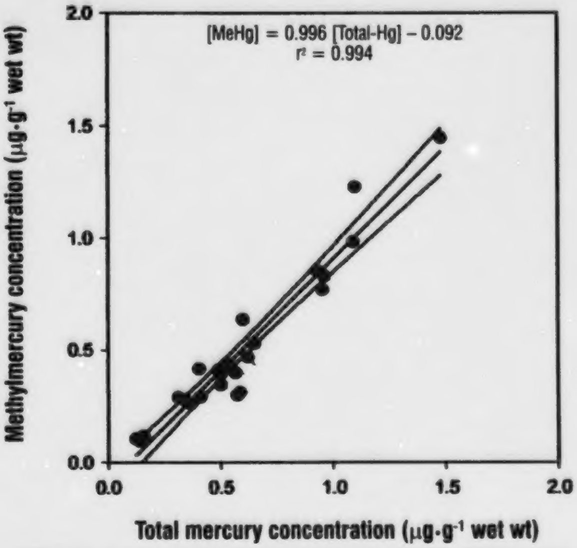
Methylmercury as a proportion of total mercury

It is often assumed that levels of total Hg in fish are a good predictor of levels of MeHg. That assumption usually holds for fish muscle but it does not hold for some organs of marine mammals. We selected 26 northern pike with a range of concentrations of total Hg in muscle from 0.123 to 1.482 $\mu g{\cdot}g^{-1}$ and analyzed them for MeHg in order to determine whether most of the Hg was in fact present as MeHg. The methods used were described by Wagemann et al. (1997). The results are shown in Figure 11. The range of MeHg concentrations found was from 0.093 to 1.444 $\mu g{\cdot}g^{-1}$, matching closely that for total Hg. The proportion of the total Hg represented by MeHg was calculated for each fish and the range was from 51.3% to 111%, with a mean of 80.1%. There was no indication of curvature in the relationship indicating that it holds over the range of concentrations tested.

Lake size and perimeter

Map data on Yukon, Northwest Territories and Nunavut lakes and rivers were obtained in MapInfo format and most of the lakes for which we have fish Hg data were found on these maps. This software permits produces and

Figure 11. Total mercury and methylmercury in muscle of northern pike (n = 26) from Little Doctor, Mahoney, Sanguez, Kelly and Willow Lakes



estimate of the lake area and lake perimeter directly from the maps. Data for some small lakes (e.g. Tsetso) or for lakes at very high latitudes were not present on the maps and so data for these are not available yet. Nonetheless there were about 40 lakes ranging in size from a few square kilometres up to Great Bear Lake (31 250 km²) for which size and perimeter data and also data on Hg levels in lake trout were available. There was no statistical relationship between Hg and either lake size or lake perimeter or between Hg and the ratio of area:perimeter.

Comparisons with geological maps

The levels of Hg in the fish have been compared with a series of geological maps available from the project leaders. Five of the lakes are in an area identified as paleozoic sedimentary rocks and the remaining two (Reade and McEwan) in an area of paleozoic-mesozoic sedimentary rocks. We cannot rigorously compare the lakes for Hg levels in the fish from all the lakes because the mix of species differed from lake to lake. However, the three northern lakes had relatively low levels in both lake trout and lake whitefish. McGill and Deep Lakes had higher levels in the whitefish and very high levels in the northern pike and walleye. The other two southern lakes, McEwan and Reade, had intermediate levels in northern pike. Comparing the regional geological maps with the levels of Hg in the fish, there are no obvious patterns in the geology that provide a confident prediction of the Hg in the fish. Several of the maps show McGill and Deep Lakes in different settings than the nearby pair of McEwan and Reade and so those differences will be explored in greater detail to see if they offer a potential explanation for the lower levels in pike and walleye from the latter pair of lakes.

On a larger scale, the mean Hg levels in lake trout in Figure 10 may be compared with the geological maps and the larger data set makes discrimination of regional geological influences even more difficult. Intuitively, geological Hg must drive at least some and possibly most of the accumulation in the fish and so the explanation will have to be sought in geological information at a finer scale than provided by the maps.

Conclusions

Of the lakes for which new data are available for 1999-2000, two of them, McGill and Deep Lakes, yielded fish with mean Hg levels over the guideline level of 0.5 $\mu g \cdot g^{-1}$ used to regulate the commercial sale of fish in Canada. The northern pike sample from Reade Lake was below the guideline level but only slightly so with a mean Hg level of 0.43 $\mu g \cdot g^{-1}$. The graph for Reade Lake (Figure 2a) shows that about one quarter of the northern pike, mostly those over 650 mm long, exceeded the guideline. Only McEwan Lake had all the mean values well below the guideline and even there the largest northern pike and walleye exceeded it.

Considering the larger set of data available for lake trout (Figure 10), the samples reported here for that species (Aubry, Colville, Belot Lakes) are in the range of previous data from northern lakes. The larger sample numbers in the current collections offer greater statistical confidence than the small sample numbers from some of the earlier collections. We have no information on year-to-year variability in Hg concentrations in the fish within a lake.

Several of the lakes have now been sampled on more than one occasion. Some instances suggest that Hg levels in fish have changed over time but others do not. There is still no obvious or consistent trend to either higher or lower concentrations. Individual lakes sometimes show apparent trends but the data overall do not suggest any regional trend.

Northern pike from northern lakes have about 80% of their total levels of Hg in muscle present as MeHg.

Comparing the Hg levels in fish from different lakes with regional geological maps did not reveal any obvious pattern of higher or lower levels associated with regional geology. It is, however, difficult to imagine that local geological settings do not influence the Hg in the fish and so geological controls on Hg in fish are probably quite local and simply not visualized at a regional scale.

There is essentially no limnological or biological information on most of these lakes. The mapping software can provide figures for lake area and lake perimeter for all lakes shown at the scale of the source maps. These factors, alone or in combination, do not appear to relate directly to the mean levels of Hg in the lake trout. However, a myriad of physical and biological factors remain to be examined in efforts to explain the levels in the fish. We have recently purchased software, called Vertical Mapper, which is intended to help develop watershed maps and possibly wetland maps from digital topographic data.

Expected Project Completion Date

This project is ongoing. It will not be completed until good descriptive models are developed that permit the accurate prediction levels of Hg in the fish without having to measure them. Until then, fish from lakes of concern to local Renewable Resource agencies will have to be analyzed.

With regard to geological influences on Hg levels in fish, it is important that this contribution be measured. If it turns out to be a major source, we cannot expect future controls on emissions of Hg to help solve the problem of high levels in fish from northern lakes. On the other hand, if local geology is not an important contributor, fish levels should improve as Hg emissions fall. It is hoped that future work with communities will allow selection of more lakes in defined local geological settings. For example, there are some lakes in southeastern Yukon near the Tintina Fault where sediments are rich in Hg (Painter et al. 1994) and fish from those lakes should be analyzed. Compilation of more detailed physical, geological and limnological information at a local scale will be required. Some of this will have to be done retrospectively for lakes for which we already have fish data. This information will help to explain some of the variation.

Logically, this project should have a marine component since marine mammals that eat fish build up very high concentrations of Hg in some organs. However, the limited existing data we have on Arctic marine fish suggests that they do not contain high levels of Hg, with the exception of Greenland sharks.

Acknowledgments

Several scientists allowed use of their data or contributed samples. Among them are Dr. Karen Kidd, Fred Tapuna, the former DFO Inspection Service (now part of the Canadian Food Inspection Agency), Mark Palmer and Pat Roach.

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Spatial Trends and Pathways of Persistent Organic Pollutants and Metals in Fish, Shellfish and Marine Mammals of Northern Labrador and Nunavik

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Abstract

The objectives of this study were to develop baseline data on persistent organic pollutants (POPs) and metals in freshwater and anadromous fish, shellfish and marine mammals that are important sources of nutrition to Inuit communities of Northern Labrador and Nunavik. 1999-2000 was the final year of the project. Successful collection of mussels (*Mytilus edulis*), sea-run Arctic char, scallops and walrus samples were made in 1999, and chemical analyses of POPs and metals in ringed seals and char collected in 1998-1999 were completed. Low concentrations of mercury, selenium and lead were found in samples of scallops from Labrador, while cadmium and arsenic levels were much higher than the other elements, especially in gut samples. Arsenic was the most prominent of the five metals determined in mussels from Nunavik. Mercury levels were low (0.02 to 0.03 $\mu g \cdot g^{-1}$ wet wt) in char collected in Labrador in 1999 and similar to our previous observations in Labrador and Nunavik. Much higher levels of mercury and selenium were found in landlocked char (at Kangiqsujuaq) than in all sea-run char from widely separated sites in Nunavik and Labrador. Mercury and selenium levels in seal liver did not differ among the five locations after adjustment for age of the animals. Percent organic Hg levels increased with age in seal muscle, from about 80% in animals 0 to 2 years of age to about 100% in adult animals. Mercury levels in walrus meat (muscle) were relatively low compared with liver and kidney. Levels

of tributyl tin in char muscle ranged from < 0.01 to 0.85 ng·g^{-1} wet wt and highest levels were found in samples from Kangirsuk (Ungava Bay region). Polychlorinated biphyenyls (PCBs) and other organochlorines were present at very low levels in mussels and Arctic char from locations in Nunavik and Labrador. In general, levels of PCBs and ΣDDT in ringed seal blubber in this study were similar to levels found in ringed seal blubber at other eastern Arctic locations.

Objectives

1. To determine spatial and temporal trends of persistent organic pollutants (POPs) and metals in freshwater and anadromous fish, shellfish and marine mammals that are important sources of nutrition to Inuit communities of Northern Labrador, Nunavik and Nunavut, for use in human health risk assessment and to contribute to discussion of the effectiveness of international actions to control POPs.
2. To develop baseline data on contaminants in marine biota from Labrador to allow comparison with those from other Canadian Arctic regions.

Description

The Nunavik/Labrador Action Plan meeting (May 1997) recommended that a comprehensive contaminants monitoring program should be developed in Nunavik and Labrador, with a special emphasis on studies in Labrador. This meeting was attended by community representatives from Labrador and Nunavik as well as by representatives of various agencies dealing with contaminant and human health issues in the North. Information gaps on contaminants in Nunavik and Nunavut were identified at the meeting. The need to determine contaminants, especially in anadromous and freshwater fish, seals and mussels was identified as very important to the residents of Nunavik and Labrador. The proceedings of the workshop were circulated to attendees in the fall of 1997 (Muir 1997).

Beginning in fall 1997, analysis of samples of ringed seal, char and blue mussel from Labrador began with the help of archived seal and char samples provided by the Department of Fisheries and Oceans. Work continued in 1998 with analysis of ringed seal, Arctic char and mussel samples collected by the Labrador Inuit Association in three Labrador communities and by residents of several communities in Nunavik. Results have been reported in two previous Northern Contaminants Program (NCP) synopsis reports (Muir et al. 1999a, Muir et al. 1999b). The initial results showed that levels of heavy metals and organochlorines in ringed seal tissues and in char muscle from Labrador were similar to levels in samples from Nunavik. Arsenic (As) was the most prominent toxic element found in mussels (1.5 to 2.2 μg·g^{-1} wet wt). Butyl tins were detected in all samples from Labrador although at relatively low levels compared to other reports for marine biota near harbours. Dibutyl tin (DBT) was the most prominent butyl tin possibly reflecting degradation of tributyl tin (TBT). Concentrations of butyl tins in seal blubber were at the low end of the range reported for sea lion livers from the coast of Japan (Kim et al. 1996); there are no previous reports for ringed seal tissues.

This report describes results for samples collected in 1999-2000 as well as some not reported previously from 1998. Collections were conducted by hunters and fishers from each community and coordinated in Labrador by the Labrador Inuit Association and in Nunavik by the Nunavik Research Centre (NvRC).

Activities in 1999-2000

Sample collection

Successful collection of mussels (*Mytilus edulis*), sea-run Arctic char, scallops and walrus samples were made as planned for 1999-2000. We were unsuccessful in obtaining bearded seal or harp seal samples. As a result, we added some unplanned analyses of seal tissues including metallothionein in liver and kidney and determination of toxaphene congeners in addition to total toxaphene, in blubber. Ringed seal blubber samples from Salluit, Kangirsuk and George River (Kangiqsualujjuaq) obtained in spring 1998 were analyzed as planned. Hunters had obtained samples of blubber, brain/cerebrum, kidney, muscle and liver plus the lower jaw for tooth aging, using a kit provided by the NvRC. All samples were shipped to the NvRC for subsampling and distribution to the analytical labs. Biological measurements on the seals and fish (e.g. age, size, sex) were made at the NvRC in Kuujjuak.

Chemical analysis

Heavy metals

Samples were analyzed for heavy metals (cadmium [Cd], mercury [Hg], lead [Pb], selenium [Se] and As) at NvRC using atomic absorption spectrometry (AAS). A subset of the samples were analyzed for organic Hg using an improved version of the original Magos method (Callum et al. 1981, Shum et al. 1979). In this method the tissue is first homogenized with Tris buffer and treated with protease enzymes to degrade proteins. Cysteine, $CuSO_4$ and acidified NaBr are added, and the mixture is extracted with toluene. The organic Hg is back extracted with thiosulfate and, after digestion, analyzed by AAS.

Metallothionein

Metallothionein (MT) was determined by M. Kwan's lab at NvRC using the silver replacement method of Scheuhammer and Cherian (1991). In brief, tissues were homogenized in cold 0.25M sucrose solution, centrifuged and then the supernatant stored at –70°C awaited for MT extraction. The supernatant was mixed with glycine buffer, silver solution in glycine buffer, and 1.5% bovine haemoglobin, type II in 30mM Tris buffer. The silver replaced all metal ions in MT molecules. After heating to precipitate proteins, except MT which is thermally stable, the mixture was centrifuged and 0.2 mL haemoglobin solution was then added to each tube. This was repeated (a total of three haemoglobin additions) and the solution centrifuged. Silver concentration in each supernatant was determined by flame AAS and the MT level was calculated from the silver concentration using a standard curve. Detection limit for the procedure was 8.3 μg MT per gram dry wt.

Butyl tins

Analysis for butyl tins was conducted by electrospray ionization tandem mass spectrometry or multiple reaction monitoring with five different channels method using electrospray ionization (MS/MS) in J. Banoub's lab. TBT, DBT and monobutyltin (MBT) were extracted from fish and mussel tissues with *n*-butanol. The butanol extract was filtered and washed with hexane to remove part of the fats and then evaporated under reduced pressure, dissolved in methanol and introduced in a flow of methanol-water into the electrospray mass spectrometer ion source. Quantification of TBT, DBT and MBT was obtained by an MS/MS method.

PCBs and other organochlorines

Polychlorinated biphyenyls (PCBs), DDT and toxaphene were determined in seal tissues, char and mussels at the National Water Research Institute (NWRI) by the National Laboratory for Environmental Testing (NLET). In brief, fish and mussel samples were homogenized and Soxhlet extracted with dichloromethane:hexane (1:1). Organochlorines were isolated by gel permeation chromatography (GPC) followed by silica gel cleanup then analyzed by gas chromatography with electron-capture detection (GC-ECD). PCBs and other organochlorines were analyzed by GC-ECD. Separation was accomplished on a HP 6890 GC using a 30 m DB-5 column with H_2 carrier gas. ΣPCBs represented the sum of 103 congeners (see listing in Table 6 footnote). Toxaphene was analyzed by low resolution GC-negative ion MS using a HP 5973 MSD in negative ion mode.

Coplanar PCBs

The toxic PCB congeners 77,126 and 169 were determined by M. Lebeuf's lab at Institut Maurice Lamontagne (IML) on subsamples of ringed seal blubber extracts (lipid removed by GPC) provided by NLET. Extracts were spiked with ^{13}C-non-*ortho* PCB surrogates by NWRI and shipped to IML, where they were subjected to chromatography on carbon columns to isolate planar PCBs prior to GC-MS/MS (ion trap) analysis for coplanar PCBs (77, 126 and 169). Mono-*ortho* PCBs (105, 114, 118, 156) were analyzed by NLET using GC-ECD.

Quality assurance

Quality assurance included the analysis of reference materials for heavy metals and organochlorines, reagent blanks and duplicate samples. The NvRC and NLET organics lab are participants in the NCP Quality Assurance Program and in the Quality Assurance for Marine Environmental Monitoring in Europe programs for heavy metals and PCBs, respectively. The NLET is certified by the Canadian Environmental Analytical Laboratory program of the Canadian Standards Association.

Statistical analyses

Mean concentrations of metals and organochlorines in char and ringed seals were compared among locations using the General Linear Models procedure in Systat (Version 9) (SPSS 1999). The analysis of covariance (ANCOVA) model included age, sex, weight or length. Results were first tested for normality using skewness and kurtosis tests. All organochlorines and metals data were sufficiently close to normal distribution that they were used without transformation. Organochlorines concentrations in ringed seals were normalized to 100% lipid. Where age or size were significant, adjusted least square means were calculated. Comparisons of least squares means from the ANCOVA were made using Tukey's (post-hoc) test.

Results

Heavy metals

Low concentrations of Hg, Se, and Pb were found in samples of scallops from Labrador (Table 1). Cd and As levels were much higher than the other elements, especially in gut samples. Muscle (meat) tissue from the scallops had relatively low levels of all five metals. Organic Hg was determined in a subset of the scallop muscle samples. All Hg in muscle was in the organic form, probably methylmercury.

Table 1. Average ($\mu g \cdot g^{-1}$ wet wt) and range of concentrations of mercury, organic mercury, selenium, cadmium, lead and arsenic in scallops from Labrador (Nain), 1999

Tissue	N		Age (years)	Total mercury	Organic mercury*	Selenium	Cadmium	Lead	Arsenic
Muscle	18	mean	19	0.03	0.03	0.19	0.95	< 0.001	0.36
		min	9	0.01	0.02	0.13	0.32	< 0.001	0.25
		max	30	0.05	0.03	0.26	2.45	0.002	0.48
Gut	18	mean	19	0.05	ND**	2.83	6.15	0.246	2.61
		min	9	0.03		1.63	3.41	0.151	1.98
		max	30	0.07		5.50	10.56	0.350	3.12
Gonad	19	mean	19	0.02	ND	0.81	1.68	ND	1.38
		min	9	0.00		0.52	0.11		0.48
		max	30	0.06		1.18	4.16		2.78

* Organic mercury using the methods of Callum et al. (1981) and Shum et al. (1979)

** Not determined

Arsenic was the most prominent of the five metals determined in blue mussels from Nunavik (Table 2). Concentrations ranged from 1.5 $\mu g \cdot g^{-1}$ wet wt in samples from Makkovik to 2.3 $\mu g \cdot g^{-1}$ wet wt in samples from Kuujjuaq. Arsenic has been previously found to be present at $\mu g \cdot g^{-1}$ levels in mussels from Nunavik (Doidge et al. 1993) but has not been routinely determined in marine or anadromous fish or shellfish from other areas of the Canadian Arctic. Hg levels were low in mussels, ranging from 0.01 to 0.03 $\mu g \cdot g^{-1}$ wet wt. Organic Hg averaged 54% of total Hg. This organic Hg is probably all in the form of methylmercury (Wagemann et al. 1997). Cd levels ranged from 0.2 to 1.1 $\mu g \cdot g^{-1}$ wet wt. Doidge et al. (1993) found similar, low $\mu g \cdot g^{-1}$ (wet wt) levels of Cd (0.5 $\mu g \cdot g^{-1}$ wet wt) and Hg (0.03 $\mu g \cdot g^{-1}$ wet wt) in blue mussels from six communities in the Hudson Bay, Hudson Strait and Ungava Bay area. There were no major differences in levels of metals between mussels collected in Nain and Makkovik and those from Nunavik.

Arsenic and Se were the most prominent of the five metals in sea-run Arctic char samples from three communities in Labrador (Table 3). There were no significant differences in concentrations (ANCOVA and Tukey's test of adjusted means) in Se or As in samples from these locations. Compared with results from samples collected in 1998 (Muir et al. 1999b), As levels were significantly higher in sea-run char at two of the four locations in Nunavik than at the three in Labrador. There has been little previous work on As in fish from northern Quebec or Labrador. Significantly lower Se levels (ANCOVA, Tukey's test) were found in char from Tasuijaq (Nunavik) compared to Labrador while other locations had similar levels. Landlocked char (Kangiqsujuaq) had significantly higher levels of Se than all sea-run char. Age and size of fish had no significant effect on Se or As levels in muscle.

Mercury levels were low (0.02-0.03 $\mu g \cdot g^{-1}$ wet wt) in char from Labrador collected in 1999 (Table 3) and similar to our previous observations in Labrador and Nunavik (Muir et al. 1999a, Muir et al. 1999b). We had previously reported undetectable levels of Cd and Pb in char muscle (< 0.001 to < 0.006 $\mu g \cdot g^{-1}$ wet wt) and these metals were again near or at detection limits. Considering the entire data

Table 2. Average ($\mu g \cdot g^{-1}$ wet wt) and range of concentrations of mercury, organic mercury, selenium, cadmium, lead and arsenic in mussels from Nunavik and Labrador, 1998-1999

Location	Total mercury	Organic mercury	Selenium	Lead	Cadmium	Arsenic
Makkovik	0.02	0.010	0.60	0.09	0.27	1.52
Nain	0.01	0.005	0.59	0.12	0.33	1.89
Kangiqsualujjuaq	0.03	0.014	0.41	0.12	0.23	2.06
Quaqtaq	0.01	0.005	0.69	0.21	1.10	1.94
Kangiqsujuaq	0.01	0.005	0.48	0.15	0.35	2.13
Kuujjuaq	0.02	0.010	0.50	0.16	0.20	2.33
Deception Bay	0.01	0.005	0.68	0.28	0.39	2.23

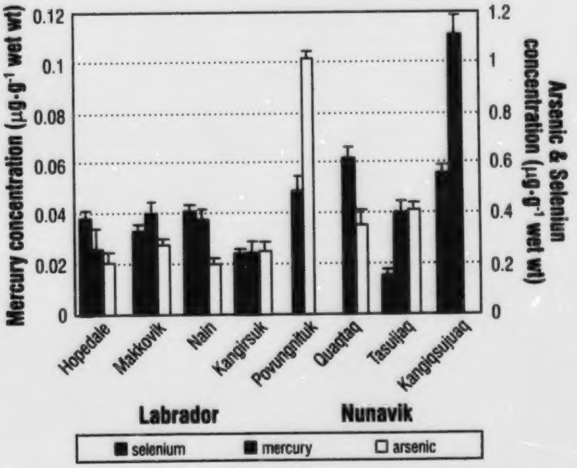
Table 3. Average ($\mu g \cdot g^{-1}$ wet wt) and range of concentrations of mercury, selenium, cadmium, lead and arsenic in arctic char from Labrador, 1999

Location	N		Length (cm)	Weight*(kg)	Age (years)	Mercury	Selenium	Cadmium	Lead	Arsenic
Nain	10	mean	48.8	1.2	7	0.03	0.38	0.001	0.001	0.18
		min	44.5	0.9	4	0.02	0.31	0.001	0.001	0.15
		max	56.5	1.6	10	0.05	0.51	0.003	0.001	0.24
Makkovik	9	mean	42.4	0.7	6	0.03	0.35	0.002	0.005	0.21
		min	37.5	0.5	3	0.02	0.29	0.001	0.001	0.16
		max	47.5	1.2	8	0.05	0.43	0.006	0.012	0.26
Hopedale	7	mean	40.6	0.9	6	0.03	0.37	0.001	0.002	0.2
		min	22.5	0.5	4	0.01	0.32	0.001	0.001	0.14
		max	53	1.7	8	0.04	0.41	0.002	0.010	0.31

* Gutted, whole fish

set (1998 and 1999) (128 samples), average Hg levels in sea-run char were similar in all samples from sites in Labrador and Nunavik, ranging from means of 0.032 to 0.040 $\mu g \cdot g^{-1}$ wet wt. Statistical analysis of the Hg results (ANCOVA, Tukey's test of adjusted means) showed that there were no significant differences in Hg levels in char among the three Labrador sites; however, one location in Nunavik (Quaqtaq) had higher levels than all other sites. Hg levels were influenced by age and size of the fish. About three-fold higher levels of Hg were found in landlocked char from Kangiqsujuaq than sea-run char. However, Se, which may exert a protective effect for Hg exposure, was also much higher in the landlocked char (Figure 1).

Figure 1. Concentrations of selenium, arsenic and mercury in Arctic char muscle collected near communities in Labrador and Nunavik*

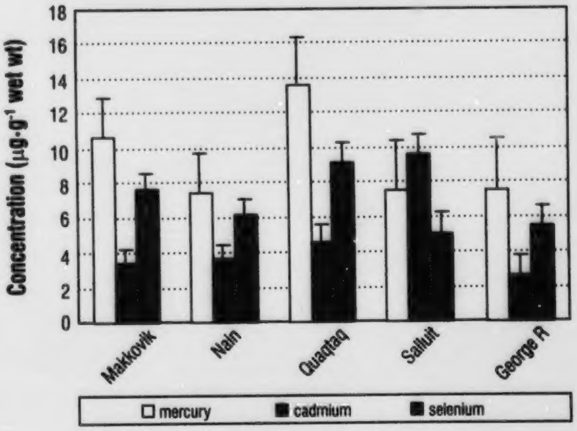


* Bars are means ± standard errors. Mercury levels are adjusted for age and length using analysis of covariance.

Results for metals in ringed seal tissues (liver, kidney, muscle and brain/cerebellum) were reported in 1999 (Muir et al. 1999b) and no further samples were analyzed. However statistical analyses were conducted to see if levels of Hg and other metals differed among locations and between Labrador and Nunavik sampling sites. Age was a significant co-variate for Hg, Se and Cd, but not As. Sex of the seals was not a significant co-variate. Hg and Se levels in seal liver did not differ among the five locations after adjustment for age (Figure 2). Cd levels were significantly higher in liver of ringed seal from Salluit than at other locations in Nunavik or Labrador (Figure 2). Arsenic levels were significantly higher at Quaqtaq than at two sites in Labrador (results tabulated in Muir et al. 1999b).

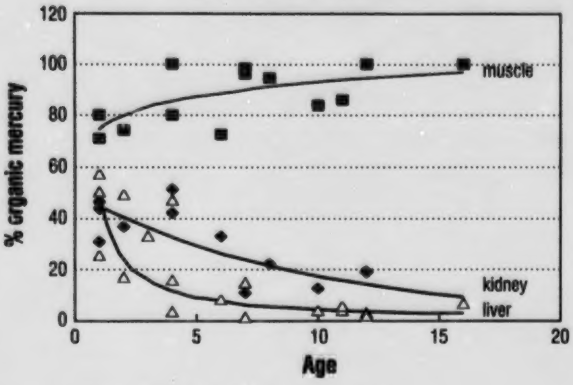
Organic Hg levels were determined in muscle, liver and kidney of 20 ringed seals from sites in Nunavik for which

Figure 2. Concentrations of selenium, cadmium and mercury in seal liver from Labrador and Nunavik*



* Bars represent age adjusted mean concentrations. Age of the seal was a significant co-variate for these elements.

Figure 3. Variation in organic mercury as a percent of total mercury in muscle, kidney and liver tissues with age of 20 ringed seals from Nunavik*



* Lines represent the best fit for the data using Excel graphics.

we previously reported total Hg (Muir et al. 1999b). The results show that organic Hg levels increased with age from about 80% of total Hg in animals 0 to 2 years of age to about 100% of total Hg in adult animals (Figure 3). Percent organic Hg declined with age in seal liver and kidney, representing less than 5% of total Hg in the oldest animals. Based on results of Wagemann (1995) and Wagemann et al. (1997), the organic Hg is likely to be entirely in the form of methylmercury.

Mercury levels in walrus meat (muscle) were relatively low compared with liver and kidney (Table 4). About 75% of Hg in muscle, 5% in liver and 20% in kidney was in the form of organic Hg, probably methylmercury (Wagemann et al. 1997). Se levels were high in all three walrus tissues while Cd concentrations were elevated only in liver and kidney. The Hg and Cd levels found in walrus tissues were comparable to those reported previously by Wagemann et al. (1996) for the same population at Inukjuak.

Metallothionein in seal tissues

MT concentrations ranged from about 50 to 600 $\mu g{\cdot}g^{-1}$ (dry wt) in ringed seal liver and from 50 to 1200 $\mu g{\cdot}g^{-1}$ in kidney (Figure 4). MT levels were strongly correlated with Cd concentrations in liver and also in kidney (for Cd concentrations $< 150\ \mu g{\cdot}g^{-1}$) (Figure 4). MT is a low-molecular weight (6.0 to 6.5 kDa), cytoplasmic protein that occurs naturally at background levels in most organisms. It is characterized by a high content of zinc (Zn), copper (Cu) and Cd, depending on availability, and an amino acid composition characterized by a high cysteinyl residue content. The induction of MT, primarily in response to Cu, Zn and Cd exposure, is a protective cellular response and MT is therefore a potential biomarker for Cd toxicity. When this protective capacity is exceeded, particularly for Cd, toxicity could occur. MT levels in seal liver were within the range found in narwhal liver by Wagemann et al (1993) but lower than the levels found in seabird liver by Sheuhammer and Cherian (1991).

Butyl tins

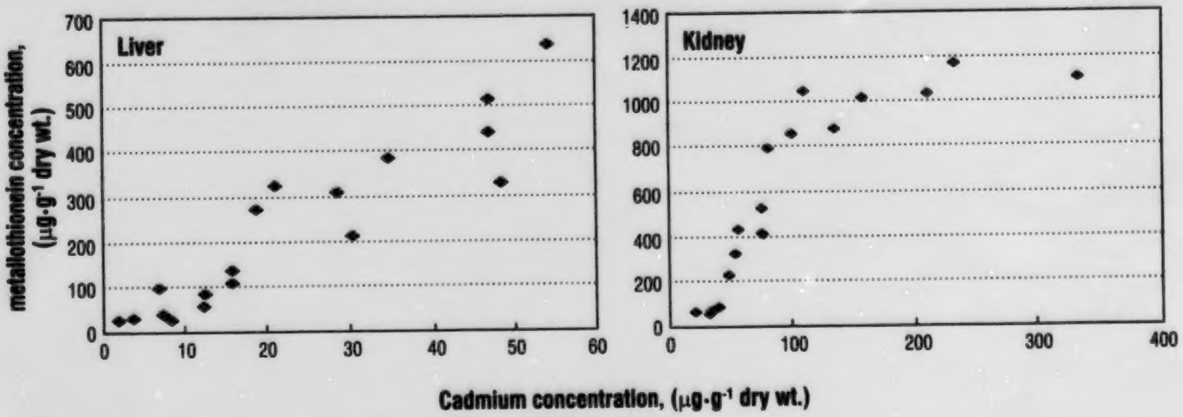
Levels of TBT in char muscle ranged from < 0.01 to 0.85 $ng{\cdot}g^{-1}$ wet wt (Table 5). Highest levels were found in samples from Kangirsuk (Ungava Bay region). ANCOVA showed that there were significant differences among locations but no effect of age. Weight was a significant co-variate. Comparison of weight adjusted means using Tukey's test showed that TBT levels in samples from Kangirsuk were significantly higher ($p < 0.05$) than other locations, while levels in char from Nain, Quaqtaq and Tasuijaq did not differ.

Table 4. Average ($\mu g{\cdot}g^{-1}$ wet wt) and range of concentrations of mercury, organic mercury, selenium, cadmium, lead and arsenic in walrus tissues from Nunavik (Inukjuaq), 1999

Tissue	N		Total mercury	Organic mercury*	Selenium	Cadmium	Lead	Arsenic
Muscle	6	mean	0.04	0.03	2.02	0.03	0.011	0.15
		min	0.03	0.02	1.38	0.01	0.006	0.09
		max	0.05	0.04	3.32	0.07	0.022	0.2
Liver	5	mean	2.64	0.12	2.59	3.32	0.098	0.27
		min	1.46	0.09	1.72	0.52	0.030	0.14
		max	3.81	0.16	3.43	8.68	0.146	0.4
Kidney	7	mean	0.31	0.06	3.85	16.49	0.184	0.22
		min	1.46	0.09	1.72	0.52	0.030	0.14
		max	3.81	0.16	3.43	8.68	0.146	0.4

* Organic mercury using the methods of Callum et al. (1981) and Shum et al. (1979)

Figure 4. Relationship between metallothionein levels and cadmium concentrations in ringed seal liver and kidney*



* The linear relationship in liver suggests that most cadmium is associated with protein in the cytoplasm.

Table 5. Concentrations of TBT ($ng \cdot g^{-1}$ wet wt) in arctic char muscle from Labrador and ringed seal blubber from Nunavik

	N	Mean	SD*	Minimum	Maximum
Kangirsuk	15	0.34**	0.36	0.01	0.85
Nain	14	0.08**	0.15	0.01	0.43
Quaqtaq	14	0.06**	0.12	0.01	0.34
Tasuijaq	15	0.05**	0.11	0.01	0.35
Kangirsuk	2	0.72***	—	0.01	1.42
Salluit	5	0.65***	0.46	0.01	1.17

* Standard deviation

** Arctic char muscle

*** Ringed seal blubber

TBT was also present in seal blubber at low $ng \cdot g^{-1}$ concentrations (Table 5). These levels were similar or slightly lower than those we reported previously in ringed seal from Labrador (Muir et al. 1997). Work is continuing on the determination of TBT in seal tissues and mussels.

Organochlorines

PCBs and other organochlorines were present at very low levels in mussels from 4 locations in Nunavik in the Ungava Bay and Hudson Strait area (Table 6). Although levels were low, PCBs were present at much higher concentrations (range of means: 7.1 to 46 $ng \cdot g^{-1}$ wet wt) in mussel tissues compared to other organochlorines. HCH isomers were the next most prominent contaminants with means ranging from 1.1 to 2.9 $ng \cdot g^{-1}$ wet wt. The greater sorption of PCBs to sedimenting particles, which are filtered by mussels, compared to most other organochlorines, especially HCH, may account for the much higher amounts of PCBs. Doidge et al. (1993) found low $ng \cdot g^{-1}$ (wet wt) levels of PCBs and organochlorine pesticides in a survey of blue mussels from six communities in Nunavik. Levels in these samples from Nunavik were higher than those found in the same species collected in fall 1997 in Labrador and reported previously (Muir et al. 1999a). Additional samples from Labrador, received late in 1999-2000, are being analyzed to obtain a better indication of prevailing levels.

Low ($ng \cdot g^{-1}$ wet wt) levels of organochlorines were detected in Arctic char muscle (+ skin) from two locations in Labrador and three locations in Nunavik (Table 7). PCBs were the most prominent contaminants averaging 11 $ng \cdot g^{-1}$ wet wt in samples from Makkovik and 31 $ng \cdot g^{-1}$ wet wt in samples from Nain. ΣPCB levels in char muscle samples from the three locations in Nunavik were intermediate in concentration, with means ranging from 14 to 21 $ng \cdot g^{-1}$ wet wt. Statistical analysis using ANCOVA and Tukey's test showed that there were no significant differences ($p > 0.05$) in ΣPCB among locations. Lipid content, length and age were not significant co-variates. However, ΣPCB levels were shown to be influenced by the sex of the fish, with males having significantly higher levels than females. This may be due to the timing of sampling near spawning time for the char. Females may have sequestered contaminants in eggs as fat is mobilized; this may lower concentrations observed in muscle.

The next most prominent groups of organochlorines in char muscle were the HCHs (hexachlorocyclohexanes) and ΣDDT (see components listed in footnote of Table 6) which were both present at low $ng \cdot g^{-1}$ levels. ΣDDT levels were higher at Nain than other locations, however using ANCOVA and Tukey's test this difference was significant only between Nain and Kangiqsujuaq ($p = 0.08$). The char from Kangiqsujuaq were landlocked. In general, these

Table 6. Concentrations of persistent organochlorine pesticides* and PCBs ($ng \cdot g^{-1}$ wet wt) in blue mussels (*Mytilus edulis*) from Nunavik**

Location	N	% Lipid	ΣCBz	ΣHCH	ΣCHL	Dieldrin/ Aldrin/ Endrin	ΣDDT	Mirex	tri-CBs	tetra-CBs	penta-CBs	hexa-CBs	hepta-CBs	octa-CBs	ΣPCB
Quaqtaq	2 pools	5.0	0.57	2.79	1.23	1.20	1.30	0.04	5.02	9.74	10.8	3.78	0.82	0.17	29
Kuujjuaq	2 pools	3	1.11	1.37	0.83	0.78	0.87	0.03	6.80	18.22	16.3	5.56	1.16	0.22	45.8
Kangiqsujuaq	2 pools	4.8	0.63	2.94	1.66	1.25	1.49	0.01	2.26	4.85	4.9	2.04	0.38	0.07	13.9
Kangiqsualujjuaq	2 pools	2	0.29	1.12	0.69	0.55	0.50	0.02	1.09	2.41	2.82	0.98	0.20	0.06	7.1

* ΣCBz = sum of 1,2,3,4-tetra, penta- and hexachlorobenzene
ΣHCH = sum of α-HCH, β-HCH and γ-HCH (except in seal muscle and liver = sum of α-HCH and γ-HCH)
ΣCHL = sum of heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, *trans*-nonachlor, *cis*-nonachlor
ΣDDT = sum *o,p'*-DDE, *o,p'*-DDD, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT

** ΣPCB = sum of 86 PCB peaks (103 congeners) (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209)

Table 7. Concentrations of persistent organochlorine pesticides and PCBs* ($ng \cdot g^{-1}$ wet wt) in Arctic char muscle from Labrador and Nunavik

Location	N		% Lipid	Age (years)	Weight (kg)	ΣCBz	ΣHCH	ΣCHL	Dieldrin/ Endrin	ΣDDT	Mirex	tri-CBs	tetra-CBs	penta-CBs	hexa-CBs	hepta-CBs	octa-CBs	ΣPCB
Makkovik	8	Mean	4.9	7	1.79	1.01	1.87	1.22	0.81	1.68	0.01	1.27	2.45	3.42	2.28	0.76	0.16	10.7
		SD**	1.2	1.2	0.54	0.29	0.32	0.62	0.19	0.69	0.00	0.98	1.86	1.69	0.73	0.20	0.03	5.1
Nain	14	Mean	6.9	8.1	1.43	1.58	3.14	1.84	1.32	2.51	0.09	3.95	8.43	8.99	6.03	2.31	0.48	30.9
		SD	2	1.2	0.32	0.56	0.92	0.64	0.58	1.27	0.23	4.58	10.7	9.43	3.16	1.22	0.24	28.6
Kangirsuk	10	Mean	8.5	7.2	2.47	1.3	1.18	1.33	1.20	1.26	0.04	3.50	6.82	6.92	2.78	0.61	0.12	21.4
		SD	2.2	1.5	3.06	0.75	0.93	0.79	0.73	0.80	0.06	2.98	5.93	5.80	1.99	0.42	0.07	17.5
Quaqtaq	8	Mean	6.1	9.2	1.46	1.31	1.14	1.26	1.12	1.36	0.04	2.76	6.04	5.69	2.49	0.61	0.14	18.2
		SD	6.1	3.1	0.77	1.16	1.25	0.98	1.04	1.13	0.04	3.34	6.58	6.09	2.27	0.47	0.11	19.1
Kangiqsujuaq	10	Mean	3.9	12	1.41	1.03	0.46	1.22	0.97	1.22	0.03	1.74	3.27	4.20	2.02	0.55	0.13	12.2
		SD	3.6	2.1	0.28	0.54	0.27	0.72	0.72	0.75	0.03	1.63	2.76	3.43	1.07	0.24	0.06	9.2

* See Table 6 for individual compounds in each organochlorine group.

** Standard deviation

levels of persistent organochlorines are similar or lower than reported in char muscle from other locations in the Canadian Arctic (Muir et al. 1997).

PCBs were the major organochlorine contaminants in the 68 samples of ringed seal blubber from Nunavik and Labrador analyzed in 1998 and 1999 and summarized in Table 8. For comparison among regions, three locations in Ungava Bay (Kuujjuaq, Kangirsuk and Kangiqsualujjuaq) were combined as were two locations on Hudson Strait (Salluit and Quaqtaq). Average concentrations of ΣPCB in blubber of males from four locations ranged from 564 to 1545 $ng \cdot g^{-1}$ wet wt and from 561 to 741 $ng \cdot g^{-1}$ wet wt in females. Highest levels in males were found in samples from Ungava Bay and Hudson Strait while highest average levels in females were from samples collected at Nain. DDT-related compounds were also prominent contaminants in ringed seal blubber with average concentrations ranging from 192 to 1344 $ng \cdot g^{-1}$ wet wt in males. Statistical analysis using ANCOVA and Tukey's test found that there were no significant differences in concentrations of ΣPCB or ΣDDT levels among the four locations for females. For males, ΣPCB, ΣDDT and ΣHCH concentrations were significantly higher ($p < 0.05$) in samples from Ungava after adjusting for age. However, there was a significant interaction of age and location, i.e. differing relationship of age of males with concentrations of the organochlorines, which may have affected the results. In general, levels of PCBs and ΣDDT in ringed seal blubber in this study have been found at other eastern Arctic locations (Muir and Addison 1998, Weis and Muir 1997). We previously found much lower levels of ΣPCB and other organochlorines in seal muscle and liver from animals from Nain (Muir et al. 1999a). Seal muscle had only slightly higher ΣPCB than levels in Arctic char and in some mussels (Muir et al. 1999b).

Toxaphene concentrations in ringed seal blubber from three Nunavik locations ranged from means of 82 to 266 $ng \cdot g^{-1}$ wet wt (Table 9). Octa and nonachlorobornanes were the major homologue groups. Congener 26 (an octachlorobornane) was the major individual toxaphene component detected. Total toxaphene levels in blubber were similar to those reported by Cleeman et al. (2000) for seals in Greenland.

Conclusions

This report provides some of the first detailed data on heavy metal and organochlorine contaminants from Labrador and the Ungava/Hudson Strait region of Nunavik. Levels of heavy metals and organochlorines in seal tissues, char muscle and mussels from Labrador were generally found to be similar to those in Nunavik. Greater differences in Hg and Se levels were found between landlocked char (at Kangiqsujuaq) and all sea-run char than among sea-run char from widely separated sites. Hg levels in seal liver did not differ among locations after adjustment for age of the animals. Organic Hg, probably in the form of methylmercury, was the major form of Hg in most tissues analyzed, i.e. in scallops, mussels, seal and walrus muscle. Future work should include direct measurements of methylmercury.

Project Completion Date

This project was not renewed in 2000-2001 after consultation with partner organizations. This report represents an interim final report. Still to be reported are results for coplanar PCBs, toxaphene, butyl tins and additional information on organochlorines in Arctic char. The final task, also not fully completed is to communicate this information to communities in the region.

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Table 8. Concentrations of persistent organochlorine pesticides and PCBs* (ng·g^{-1} wet wt) in ringed seal blubber from Labrador and Nunavik

Location**	N	Sex		ΣCBz	ΣHCH	ΣCHL	Dieldrin/Endrin	ΣDDT	Mirex	tri-CBs	tetra-CBs	penta-CBs	hexa-CBs	hepta-CBs	octa-CBs	nona/deca-CBs	ΣPCB
Ungava Bay (3 locations)	3	M	Mean	16.8	130	307	76.3	734	7.8	44.1	87.3	272	454	144	34.7	2.0	1040
			SD***	6.9	48.8	210	32.3	595	8.8	27.3	64.3	205	438	163	40.2	2.3	943
	9	F	mean	13.2	135	156	41.1	273	5.7	35.5	56.0	136	219	83.8	24.0	2.4	561
			SD	3.9	61.7	87.1	13.6	164	3.2	16.7	28.3	65.6	115	50.6	14.3	2.1	295
	8	Juv	mean	19.3	190	174	91.4	244	2.1	40.7	70.6	169	157	37.0	7.6	0.4	483
			SD	6.7	61.8	69.0	49.4	112	0.8	17.9	29.9	73.2	49.7	14.6	3.1	0.2	189
Hudson Strait (2 locations)	9	M	mean	20.6	170	402	101.6	1344	16.6	50.2	115	404	699	213	56.6	4.4	1545
			SD	27.3	126	673	145.5	2935	40.0	51.7	158	591	1347	456	127	10.6	2749
	8	F	mean	10.9	111	199	50.1	424	6.1	27.1	50.1	169	272	92.0	25.1	2.2	643
			SD	5.3	38.4	179	25.3	433	5.1	19.4	36.4	132	281	99.5	24.4	1.8	603
	11	Juv	mean	12	130	200	57.5	449	3.3	40.1	64.4	197	242	61.1	14.0	0.9	632
			SD	7.3	70.4	157	32.6	543	2.3	45.2	53.1	201	227	51.5	11.6	0.7	614
Nain	5	M	mean	9.9	86.6	104	31.5	192	2.0	22.1	45.4	126	236	105	26.6	1.1	564
			SD	6	27.3	47.0	11.5	118	1.3	8.2	20.9	52.4	115	62.0	17.0	0.8	278
	5	F	mean	10.1	102	102	25.7	164	2.4	25.8	49.5	136	304	177	44.0	1.5	741
			SD	6.2	47.3	68.6	10.8	102	1.5	16.1	32.6	88.9	217	137	29.6	0.8	523
Makkovik	9	M	mean	9.6	85.3	109	33.3	209	1.6	20.2	39.8	102	204	84.2	18.8	0.7	472
			SD	4.3	62.9	47.5	30.3	63.4	0.7	8.7	16.2	33.3	56.5	27.6	5.1	0.2	149
		F	mean	7.9	104	146	69.0	304	8.9	16.2	31.6	96.8	274	188	59.8	4.4	674

* See Table 6 for individual compounds in each organochlorine group.

** Ungava locations are Kuujjuaq, Kangsiruk and Kangiqsualujjuaq; Hudson Strait locations are Salluit and Quaqtaq

***Standard deviation

Table 9. Concentrations of toxaphene, toxaphene homolog groups* and major congeners ($ng{\cdot}g^{-1}$) in ringed seal blubber from Nunavik**

Location	Sex		Age (years)	N	Total Toxaphene	Hexa	Hepta	Octa	Nona	P26	P50	P62
Kuujjuaq	F	Mean	1.7	3	81.9	2.1	23.5	46.0	10.3	9.4	1.5	<0.1
		SD***	2.0		67.6	2.0	19.1	37.6	9.1	6.9	1.2	—
Kuujjuaq	M	mean	2.8	3	138	14.1	58.5	62.3	2.6	22.1	3.6	0.8
		SD	3.6		87.5	15.7	35.9	62.2	3.2	22.4	3.4	1.3
Quaqtaq	F	mean	7.0	4	266	6.2	77.2	141	41.4	31.8	6.6	1.3
		SD	4.3		250	5.8	83.9	123	39.4	25.1	5.1	2.1
Quaqtaq	M	mean	4.8	5	265	6.5	75.1	144	39.5	31.5	6.1	1.6
		SD	4.1		199	6.5	59.8	106	28.1	19.8	3.9	1.8
Kangiqsualujjuaq	F	mean	6.4	6	91.6	2.1	26.4	49.5	13.9	14.7	2.8	0.5
		SD	5.1		51.8	0.5	16.3	26.0	9.2	8.3	1.9	0.6

* Homologs = hexa-, hepta-, octa- and nonachlorobornanes quantified by GC-ECNIMS with a single response factor.

** Congeners = P26, P50 and P62 quantified with authentic standards.

***Standard deviation

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Temporal Trends of Persistent Organic Pollutants and Metals in Landlocked Char

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Abstract

The objectives of this study are to determine temporal trends of persistent organic pollutants and metals, especially mercury, in landlocked Arctic char in Char Lake and Resolute Lake, Nunavut, by analysis of annual sample collections, to investigate factors influencing contaminant levels in landlocked char such as the influence of sampling time, water temperature and diet, and to provide this information on a timely basis to the community of Qausuittuq (Resolute). This is the first report on this study which began in April 1999. Concentrations of mercury and other metallic elements in landlocked Arctic char from Resolute and Char Lakes were similar or slightly lower than levels in char from two other lakes in the region. There has been no significant change in mercury concentrations in char from Char Lake over the 7-year period. Persistent organochlorines were measured in Arctic char from Resolute Lake collected in 1997. Levels of polychlorinated biphenyls (PCBs) and DDT-related compounds were much lower in char from Resolute Lake compared with results from char collected in nearby Char Lake in 1993. The most likely reason for higher levels of PCBs and DDT compounds in the fish from Char Lake is that the ones we sampled in 1993 had a cannibalistic feeding habit and are feeding at a higher trophic level than those from Resolute Lake. Although levels of organochlorines in char are higher in Char Lake than the other lakes we conclude that this is not due to any recent contamination. The similar levels of heavy metals in char muscle from Char Lake and other lakes also suggests no unusual impact on the lake of the nearby Resolute airport.

Objectives

1. To determine temporal trends of persistent organic pollutants (POPs) and metals, especially mercury

(Hg), in landlocked Arctic char in Char Lake and Resolute Lake by analysis of annual sample collections.

2. To investigate factors influencing contaminant levels in landlocked char such as the influence of sampling time, water temperature and diet.
3. To provide this information on a timely basis to the community of Qausuittuq (Resolute).

Description

The Northern Contaminants Program (NCP) "Blueprint for Monitoring of Health of Arctic Peoples and Ecosystems and the Effectiveness of International Controls" (Northern Contaminants Program 1999) recommended that freshwater fish be included in a comprehensive Arctic contaminants biomonitoring program. The rationale for this is that freshwater organisms are directly exposed to atmospheric contaminants deposited on snow or directly into lake surfaces or lake watersheds. Fish accumulate contaminants and metals as they pass water over their gills and from their diet. Unlike mammals, fish have limited capacity to degrade most POPs and excrete many of these chemicals very slowly. Fish are therefore good indicators of freshwater contamination by POPs and metals.

The assessment of contaminants conducted by the NCP (Jensen et al. 1997, Muir et al. 1997) found that there was limited information on temporal trends in fish. Temporal trends in fish were available only for burbot liver from the Mackenzie River at Fort Good Hope, Northwest Territories, from the mid-1980s to 1994 and for round and lake whitefish from Lake Laberge, Yukon, from the 1970s to the 1990s. No temporal trends were available for fishes in Nunavut or in the high Arctic.

Temporal trend information was more complete for marine mammals and seabirds (Muir et al. 1997) and the NCP is currently funding additional work on temporal trends in seals and beluga (Stern 1999) and seabirds eggs (Braune 1999). While valuable information has been learned from temporal trend studies with seabird eggs and mammals, changes over time are difficult to discern over the 1 to 5 year period typically funded by the NCP, due to long half-lives of some contaminants in the animals and high rates of transfer from mothers to offspring. Furthermore, while levels of some POPs have declined in air, there is little evidence for changes in contaminants in seawater flowing through the Canadian Arctic archipelago. For all of the above reasons we can expect a relatively slow response of marine biota to changes in contaminants in the atmosphere.

Small lakes in the high Arctic are replenished annually with snowmelt runoff and direct precipitation, which represent significant fractions of their water budgets. Declining concentrations of POPs, or increasing levels of previously unstudied POPs, in air and precipitation should be reflected relatively quickly in changes in levels in food webs and top predator fishes, compared to the vast marine environment. We know this to be the case from the sedimentary record of POPs and Hg in small Arctic lakes (Muir et al. 1996, Lockhart et al. 1995).

Small high Arctic lakes also show major changes in water temperature during the short open water period of the Arctic summer. Because temperature affects contaminant uptake due to increased gill ventilation by fish, there may be seasonal differences in contaminant levels. Other factors such as increased lipids from higher productivity during summer may also influence levels of contaminants, especially POPs in fish. Köck et al. (1999) have found increased accumulation of cadmium by char in Resolute Lake over the summer period. Köck et al. (1996) have previously shown that lake water temperature was a major factor in elevated cadmium and lead accumulation by landlocked char in high mountain lakes in Austria.

The value of annual long-term sampling of fish for measurement of temporal trends of contaminants was well illustrated by studies in Sweden. Arctic char have been collected since 1980 in Lake Abiskojaure, 200 km north of the Arctic circle, and northern pike were collected from Lake Storvindeln, a forest lake in northern Sweden since 1978. Samples were collected annually and were selected for consistency in sex, age, size and sampling season. Each annual sample at a given location was represented by 10 to 25 specimens, thus within-year variation could be estimated. Declines in levels of ΣDDT (DDT + DDD + DDE), PCBs and hexachlorobenzene were noted in fish from the Arctic locations (Olsson and Reutergårdh 1986, DeMarch et al. 1998). The char data provided a more accurate temporal trend than pike muscle since the char has a higher concentration of fat. HCH (hexachlorocyclohexane) isomers showed no significant declines in fish muscle over a 24-year period. Because of annual sampling, the Swedish program was also able to detect increases in DDT inputs due to the use of DDT in East Germany in 1984 (Bignert et al. 1993).

Char and Resolute Lakes near Resolute are ideal sites for study of temporal trends of POPs and metals in fishes. These lakes ares well studied limnologically and the food chain in Char Lake has been well documented (Hobson and Welch 1995). Char Lake also serves as the water supply for the town of Qausuittuq and it is a relatively undisturbed lake with a stable landlocked Arctic char population. Char from Char Lake collected by H. Welch in 1993 have been previously analyzed for organochlorine contaminants and metals (Muir and Lockhart 1996, Muir et al. 1996). Levels of PCBs and total DDT in char muscle (plus skin) were relatively high compared to other landlocked or sea run char (Muir et al. 1996). Other persistent organochlorines (e.g. chlordane, toxaphene) in Char lake char were at levels similar to other Canadian Arctic locations. The higher levels of DDT and PCBs may represent local contamination but also could be due to trophic position of the char that were sampled. Nearby Resolute Lake is also relatively undisturbed although it is downstream from Meretta Lake which receives sewage effluent from the town of 250 permanent residents. Contaminants have also been measured in landlocked char from Amituk Lake, north of Resolute, on Cornwallis Island (Muir and Lockhart 1994).

Activities in 1999-2000

Sample collection

This project received NCP funding for the first time in 1999-2000. Samples collected in 1997 and 1998 from Resolute Lake were distributed to J. Reist and D. Muir. These previous collections were funded entirely by a grant from the Austrian Ministry of Science to Dr. Guenter Köck (University of Innsbruck). Dr. Köck collected char at approximately 30-day intervals: prior to ice-breakup in July, in mid-August and again in mid-September each year from Resolute Lake. Arctic char were collected from Char and Resolute Lakes in the summer of 1999 by Dr. Köck and his students, working with Debbie Iqaluk of Resolute, using either hand methods or gill netting. Unfortunately only three char could be obtained from Char Lake in 1999, although 20 fish were collected from Resolute Lake. Additional collections of landlocked char were made at Boomerang Lake on Somerset Island and Sapphire Lake on Devon Island. Samples were frozen in Resolute and then shipped to the National Water Research Institute (NWRI), Burlington, Ontario, and stored at −20°C until analysis. Samples collected in 1997 were dissected in J. Reist's laboratory and distributed to NWRI.

Chemical analysis

Metals

Arctic char muscle was subsampled and acid-digested using high pressure microwave. The digest was then analyzed for Hg by cold vapour atomic absorption spectrometry (CVAAS) and for 22 elements using inductively coupled plasma-low resolution mass spectrometry (ICP-MS).

Organochlorines

Char muscle samples were homogenized, mixed with pre-cleaned Na_2SO_4 and Soxhlet extracted with dichloromethane:hexane (1:1). Organochlorines were isolated by gel permeation chromatography followed by silica gel cleanup. The chromatography on silica gel was used to separate PCBs and *p,p'*-DDE from more chlordane and toxaphene components, *p,p'*-DDT and HCH isomers. The two fractions from the silica gel column were then analyzed by gas chromatography (GC) with electron-capture detection. Separation was accomplished on a HP 6890 GC using a 30 m DB-5 column with H_2 carrier gas. ΣPCBs represented the sum of 103 congeners. Toxaphene was analyzed by low resolution GC-negative ion MS using a HP 5973 MSD in negative ion mode.

Quality assurance

Quality assurance included the analysis of certified reference materials for heavy metals and organochlorines, reagent blanks and duplicate samples. The National Laboratory for Environmental Testing (NLET) organics and metals labs are participants in the NCP Quality Assurance Program. The NLET organics lab is a participant in the Quality Assurance for Marine Environmental Monitoring in Europe programs for PCBs and toxaphene. NLET is certified by the Canadian Environmental Analytical Laboratory program of the Canadian Standards Association

Results

Concentrations of Hg and other metallic elements in landlocked Arctic char from Resolute and Char Lakes were similar to or slightly lower than levels in other lakes in the region (Table 1).

Statistical analysis showed that concentration data for most elements in char muscle from Resolute, Boomerang and Sapphire Lakes were normally distributed. We therefore reported arithmetic means and conducted correlation analysis with untransformed data. Hg was strongly correlated with fish length and weight ($p < 0.05$) in all four lakes. Hg was also significantly correlated with thallium, rubidium and selenium ($p < 0.05$) in all lakes, but not consistently with any of the other 18 other elements we measured.

Concentrations of Hg in landlocked char from Resolute and Char Lakes previously reported in samples from 1992 (Muir and Lockhart 1994) are compared with the more recent samples in Figure 1. There appears to have been no significant change in Hg concentrations over the 7-year period. This is interesting because Hg concentrations increased in seabirds and marine mammals from the

Table 1. Mean and range of concentrations of mercury and other major elements* detected in arctic char muscle ($\mu g \cdot g^{-1}$ wet wt) from lakes in the Resolute/central Arctic archipelago area

Lake		N	weight (g)	As	Cd	Cr	Ni	Pb	Rb	Tl	V	Se	Hg
Resolute 1999	Mean	10	507	0.109	0.002	0.166	0.035	0.008	0.150	0.009	0.039	0.728	0.172
	Min		359	0.030	0.001	0.130	0.020	0.003	0.087	0.004	0.028	0.660	0.07
	Max		842	0.150	0.002	0.200	0.070	0.015	0.196	0.018	0.049	0.810	0.262
Resolute 1997	Mean	10	305	0.064	0.005	0.101	0.045	0.008	0.139	0.007	0.011	0.649	0.2
	Min		210	0.010	0.001	0.050	0.010	0.002	0.100	0.003	0.004	0.510	0.102
	Max		434	0.110	0.019	0.310	0.160	0.020	0.166	0.017	0.019	0.780	0.426
Boomerang 1999	Mean	11	950	0.072	0.001	0.155	0.024	0.008	0.115	0.007	0.035	0.765	0.314
	Min		362	0.010	0.001	0.140	0.010	0.004	0.038	0.002	0.030	0.620	0.104
	Max		1881	0.110	0.002	0.170	0.040	0.013	0.210	0.010	0.041	0.940	0.521
Sapphire 1999	Mean	8	864	0.046	0.002	0.135	0.025	0.007	0.360	0.016	0.031	0.670	0.291
	Min		300	0.030	0.001	0.110	0.010	0.003	0.269	0.008	0.025	0.610	0.116
	Max		3465	0.070	0.005	0.170	0.050	0.012	0.454	0.027	0.040	0.740	0.577
Char 1999	Mean	3	201	0.047	0.002	0.113	0.017	0.007	0.089	0.004	0.026	0.487	0.215
	Min		40	0.030	0.001	0.110	0.010	0.006	0.058	0.002	0.023	0.440	0.09
	Max		346	0.060	0.003	0.120	0.030	0.009	0.115	0.005	0.029	0.560	0.305

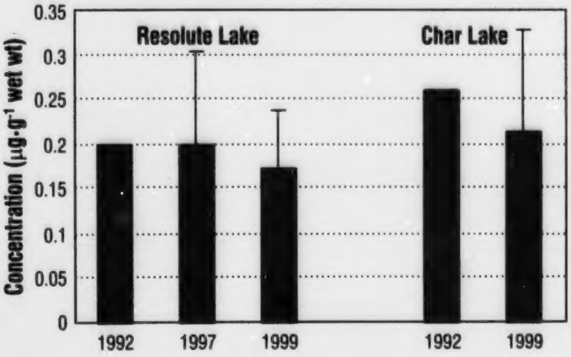
* As, arsenic; Cd, cadmium; Cr, chromium; Ni, nickel; Pb, lead; Rb, rubidium; Tl, thallium; V, vanadium; Se, selenium; Hg, mercury

1970s to the mid-1990s (Braune 1999, Wagemann et al. 1996).

Temporal trends of cadmium in char from Resolute Lake were studied in 1997 and 1998 by sampling three times during the open water season. Results showed that cadmium levels in char increased over the summer corresponding to increased water temperature. Higher cadmium in liver in 1998 compared to 1997 coincides with higher water temperatures in August 1998 (2°C in 1997, 8°C in 1998) (Figure 2). The mathematical relationship between lake temperature and metal uptake was similar in high Arctic lakes and previously studied Austrian high mountain lakes, confirming water temperature to be a key factor in metal accumulation in char (Köck et al. 1996).

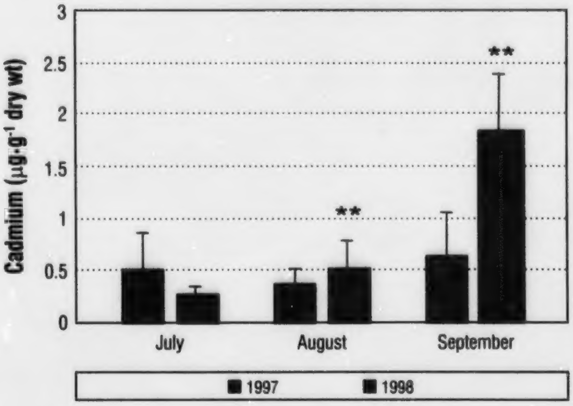
Persistent organochlorines were measured in Arctic char from Resolute Lake collected in 1997. Concentrations of major organochlorines groups are compared in Table 1 with results from Char Lake in 1993. Also included are

Figure 1. Mercury in arctic char from Resolute and Char lakes, 1992-1999*



* Bars represent mean ± standard deviation.

Figure 2. Concentrations of cadmium in liver of landlocked char from Resolute Lake 1997 and 1998*



* Bars represent mean ± standard deviation.

** Indicates significant difference between 1997 and 1998.

Table 2. Concentrations of major organochlorine chemical groups* in landlocked char from Cornwallis Island

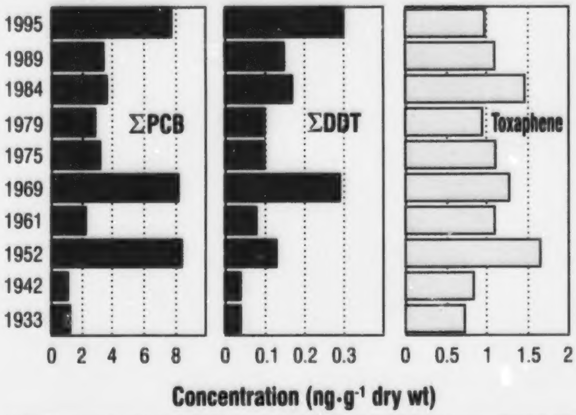
Lake	year	N	Lipid (%)	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB
Resolute	1997	10	5.2	1.2 ± 0.37	0.5 ± 0.21	4.1 ± 2.2	5.3 ± 3.6	98 ± 67
Char	1993	5	4.3	3.1 ± 1.6	2.64 ± 1.4	16 ± 6.9	114 ± 53	290 ± 120
Amituk	1992	12	4	5.9 ± 2.6	2.05 ± 1.1	42 ± 21	28 ± 14	78 ± 39

* ΣCBz = sum of tetra, penta and hexachlorobenzene, ΣHCH = sum of α, β and γ-isomers, ΣCHL = sum of *cis*- and *trans* chlordane and nonachlor, heptachlor, heptachlor epoxide and oxychlordane, ΣDDT = sum of *o,p*-DDE, -DDD, -DDT, *p,p'*-DDE, -DDD and -DDT. ΣPCB = sum of 85 PCB congener or co-eluting congener peaks

results from landlocked char in Amituk Lake (Muir and Lockhart 1994). Char in Resolute Lake had similar levels and proportions of PCBs but much lower levels of ΣDDT, ΣCHL and ΣHCH than those in Amituk Lake from 1992. The char from Char Lake from 1993 had higher levels of more bioaccumulative organochlorines such as PCBs and *p,p'*-DDE than fish from either Amituk or Resolute Lakes (Table 2). The most likely reason for higher levels of the more bioaccumulative compounds is that the char have a behavioural dichotomy wherein some individuals at a certain "escape size" adapt to a cannibalistic feeding habit, while other individuals may not. Thus some individuals are feeding at a higher trophic level. It may be possible to discern this using stable isotopes of nitrogen which increase with trophic level. Measurement of δ^{15}N is currently underway at NWRI but we have no samples from 1993 for comparison. The lower levels of ΣHCH, ΣCHL and ΣDDT in Resolute Lake char from 1997 than in samples from the early 1990s in Amituk or Char Lakes may reflect a temporal trend but further work is needed to confirm this. The issue may be clarified with completion of the analysis of organochlorines in samples from 1999 and 2000.

The question of possible contamination of Char Lake by organochlorines was raised in the *Canadian Arctic Assessment Report* (Jensen et al. 1997). Char Lake is next to the airport at Resolute and also serves as the town's drinking water supply. Contamination by organochlorines was investigated by collection and analysis of a sediment core from the lake in 1997. The sediment core (collected from Char Lake by J. Smol, Queens' University, and M. Douglas, University of Toronto) was dated at NWRI and analyzed for PCBs and organochlorine pesticides including toxaphene to determine historical inputs of organochlorines to the lake. This latter work is funded by a Toxic Substances Research Initiative project (Muir et al. 1999). Results show relatively low levels of PCBs, DDT, toxaphene and other pesticides (Figure 3). There is no indication of any unusually high contamination of Char Lake sediments by PCBs or DDT although slightly higher levels in the 1950s and 1960s might be related to DDT use for insect control or local PCB use.

Figure 3. Concentrations of ΣPCB, ΣDDT and toxaphene in dated sediments from Char lake



Conclusions

This is the first report on what is intended as a long-term study of contaminant levels in landlocked Arctic char in the Resolute Bay area. Results show that levels of Hg and metallic elements in char from Char and Resolute lakes are relatively similar to those in other lakes in the region. The preliminary results suggest no change in concentrations of Hg over the 7-year period. Further annual sampling will help to strengthen this conclusion. Organochlorine levels in char from Resolute Lake were lower than in char from Char Lake collected in 1993 but this may be related to the sampling of "cannibal" char in Char Lake. Further analysis of archived fish would help resolve this issue. Although levels of organochlorines are higher in Char Lake than the other lakes we conclude that this is not due to any recent contamination. The similar levels of heavy metals in char muscle from Char Lake and other lakes also suggests no unusual impact of the airport on the lake.

Expected Project Completion Date

Unknown

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Effects of Persistent Organic Pollutants on Polar Bears

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Abstract

A polar bear immunotoxicology project was initiated as an extension of research carried out by Norwegian Scientists in Svalbard in 1998 to polar bears from western Hudson Bay in 1999. The research includes studying the relationship between immunoglobulin levels and polychlorinated biphenyl (PCB) exposure in polar bears, the effect of high PCB exposure on the ability of the bears to produce antibodies to various vaccines, the relationship between PCB exposure and the lymphocyte proliferation in polar bears, and the effect of in vitro exposure of lymphocytes to PCB congeners. The experimental work is completed, but data analysis has only begun and there few results to report at this time. An interlaboratory calibration of analytical methods for analysis of organochlorine in plasma was carried out between the National Wildlife Research Centre and the Norwegian laboratory. Although there were some discrepancies, agreement was excellent for the major persistent organic pollutants

A method of extracting hydroxylated metabolites in plasma for analyses was developed and validated using polar bear plasma as a test tissue. A major phenolic compound in polar bear plasma was identified as 4-hydroxyheptachlorostyrene, which is likely a metabolite of octachlorostyrene. Seventy-one polar bear plasma samples were analyzed for hydroxylated metabolites of PCBs (OH-PCBs), other halogenated phenolic compounds as well as PCBs, chlordanes and other organochlorine compounds. Total OH-PCBs were equal to and generally higher concentration than total PCBs in all samples analyzed, indicating the importance of OH-PCBs in plasma. Chemical residue data was analyzed by principal component analysis. The analysis produced four major components which were closely associated with different classes of contaminants: persistent PCBs, OH-PCBs, non-persistent PCBs and

chlordanes, in decreasing order of percent variance explained. Vitamin A concentrations in the plasma were inversely correlated with PC1 which is associated with persistent PCB exposure. T4 binding index of the plasma, a measure of binding capacity of the plasma for the prohormone thyroxine, was inversely correlated with PC2, which is associated with OH-PCBs, indicating possible disruption of thyroid hormone transport in the polar bear.

Rate of formation of eight different metabolites of testosterone were studied using microsomes from 16 polar bears which had been prepared in 1993 and thoroughly characterized as to CYP1A and CYP2B enzyme concentrations, 7-ethoxyresorufin O-deethylase, 7-pentoxyresorufin O-deethylase and 7-benzyloxyresorufin O-deethylase activity, and organochlorine content of the liver. The profile of metabolites was similar to that of sheep, cattle and goats, but different from that of rats. None of the metabolites appeared to be associated with dioxin-like enzyme activity (CYP1A), however, the activities correlated in a complicated fashion with a number of PCB congeners, including the multi-ortho congeners, suggesting the possibility of CYP2B and CYP3A involvement. Further statistical analysis is required before conclusions can be drawn.

Studies of metabolism of individual organochlorines, including PCB congeners, proved difficult to set up. Optimization of conditions has now been achieved for CB47, CB52, CB77, α-HCH, β-HCH and *trans*-chlordane. This research will continue in 2000-2001, including enantio-selective metabolism of α-HCH and *trans*-chlordane, and enanto-selective formation of oxychlordane.

Objectives

Long-term

- To determine the effects, at the individual and population levels, of persistent toxic organochlorines (OCs) and their metabolites in the polar bear.

Short-term

1. In collaboration with the Canadian Wildlife Service, Prairie and Northern Region, the Norwegian Polar Institute and the Norwegian Veterinary Institute, determine the immunotoxic effects of polychlorinated biphenyls (PCBs) and other OCs in polar bears throughout a gradient of exposure (Hudson Bay, low; Svalbard, high).
2. Determine the effects of hydroxy-PCBs (OH-PCBs) on circulating thyroid hormone and vitamin A concentrations.
3. Determine the endocrine disrupting effect of persistent organic pollutants (POPs) on testosterone and PCB metabolite profiles by in vitro metabolism studies using polar bear liver microsomes.

Introduction

It has been well documented that the polar bear is among the most highly OC-contaminated of Arctic mammals, due to its almost exclusive diet of ringed seal. Monitoring of polar bear tissues for both OCs and heavy metals has been an important part of the Northern Contaminants Program (NCP) since its inception, as well as the international Arctic Monitoring and Assessment Programme (AMAP). Results up to 1996 are extensively reported (Jensen et al. 1997, Arctic Monitoring and Assessment Programme 1997). Subsequent NCP reports (Norstrom 1999a, Norstrom 1999b) contained analysis of: geographical, age and sex differences in PCB and chlordane accumulation in polar bears; effect of seasonal fasting on OC concentrations in mothers and cubs; relationship between OC concentrations in milk and the female's adipose tissue; evidence that females that lost cubs during their first year of life had statistically higher concentrations of all OCs than females that kept their cubs; indication that body burdens of PCBs and chlordanes did not change significantly during denning of pregnant females; and preliminary identification and quantification of OH-PCB metabolites in blood and their possible effect on thyroid hormone levels.

The project shifted emphasis from monitoring to effects in 1999-2000, in keeping with the evolving research priorities of the NCP and AMAP. Norwegian scientists had already begun a study on immunotoxicology and immunosuppression in polar bears in Svalbard in 1998, and developed extensive expertise. A project was therefore initiated to have the Norwegian scientists repeat the study with bears from western Hudson Bay (Cape Churchill) in 1999, in collaboration with Canadian scientists.

In addition to the immunotoxicology studies, research continued in collaboration with Carleton University on the identification and effects of OH-PCB metabolites on thyroid hormone and vitamin A levels in polar bear blood. A new study was initiated with the University of British Columbia on the possible effect of PCBs and other OCs on the in vitro metabolism of testosterone, and rate of PCB congener metabolism, using archived microsomes from 16 polar bears collected in 1993. EROD (7-ethoxyresorufin O-deethylase) enzyme activity, cytochrome P450 1A and 2B enzyme concentrations, and analysis of the complete range of OCs, including dioxins and furans, had already been obtained for these samples (Letcher et al. 1996). This allowed an investigation of the correlation of testosterone and PCB metabolism rates with a wide range of biochemical and chemical parameters.

Activities in 1999-2000

Four hypotheses were being tested:

1. The immune system of polar bears with the highest PCB concentrations is compromised.
2. OH-PCB metabolites decrease the circulating concentrations of T4 (thyroid hormone) and retinol (vitamin A). Interference with the homeostasis of these compounds affects growth and development.
3. The metabolite profile of testosterone in polar bears is affected by increased PCB exposure.
4. The rate of formation of OH-PCB metabolites in polar bears is determined by the chlorine substitution pattern and degree of exposure to PCBs (Ph.D. research, C. Sandau, Carleton University).

Immunotoxicology

The aim of the project is to assess the immunological impact of high PCB exposure in polar bears. By using specific immune function tests and induction of protective antibodies, we will obtain information on how PCBs influence the resistance to infection of polar bears. The study consisted of four parts: i) determining the relationship between immunoglobulin levels and PCB exposure in polar bears; ii) determining whether or not high PCB exposure decreases the ability to produce antibodies in polar bears; iii) determining the relationship between PCB exposure and lymphocyte proliferation in polar bears; and iv) determining the effects of in vitro exposure of lymphocytes to PCB congeners.

Between August 29 and September 4, 1999, 35 free-ranging male polar bears were captured in Manitoba along the western coast of Hudson Bay (I. Stirling and N. Lunn). At the time of capture and handling, each animal was immunized with herpes-, reo-, and influenza viruses, tetanus toxoid, and keyhole limpet cyanin (KLH) via the injection of the following vaccines: i) Prevacun7 FT (Hoechst Veterinär GmbH), Lot no. 050011B, 2 mL, intra-muscular; ii) Resequin7 plus (Hoechst Veterinär GmbH), Lot no. 13021, 2 mL, intra-muscular; and iii) KLH (Sigma), Lot no. 018H4835, 2 mL, subcutaneous.

Blood was collected and frozen plasma samples were shipped to the Norwegian School of Veterinary Science (NSVS) for subsequent analysis. To aid in their recapture, a VHF radio transmitter was placed in one ear of each of 32 of 35 of these male bears. Between October 6-10, 1999, 30 of the 35 immunized male polar bears were recaptured. The mean capture-recapture interval was 36.3 ± 0.5 days (range: 32 to 40 days). At the time of recapture, the VHF radio transmitter was removed and approximately 215 mL of whole blood was collected from each animal. Lymphocyte proliferation experiments were carried out using fresh blood samples from recaptured bears. Sampling of 25 feces collections was carried out to try to characterize normal fecal microflora of the Hudson Bay bears as a general health control.

Lymphocyte proliferation tests were successfully carried out in the laboratory at Churchill Northern Studies Centre on 30 polar bears using both mitogen (phytohemagglutinin [PHA], pokeweed mitogen [PWM], concanavalin A [ConA], mycobacteria antigen [PPD] and lipopolysaccharid [LPS]) and antigen-induced (KLH, tetanus toxoid) lymphocyte proliferation (H.J. Larsen). In addition, the effect of four PCB congeners on the ConA-induced lymphocyte proliferation in vitro was assessed. Hematology (WBC, RBC, HCT, Hb, diff. counting), serology and determination of immunoglobulin G (IgG) concentrations were done in Norway. Serology included the measurement of neutralizing antibodies against equine herpes virus (EHV), equine influenza virus (EIV), reovirus, protecting antibodies against tetanus toxoid, antibodies against KLH and *Pasteurella* spp.

Plasma from the recaptured bears was analyzed for 24 PCB congeners, DDT, DDE, dieldrin, chlorobenzenes, octachlorostyrene and 11 chlordane-related compounds at the National Wildlife Research Centre (NWRC) (S. Newson, C. Sandau). The extraction and clean-up method used was the same as for OH-PCB analysis (see below) in order to maintain consistency and comparability of data. Lipids were determined by a colorimetric method, which has been proven to give comparable results to exhaustive total lipid extraction. Chemical analysis of the initial capture blood samples for PCBs and OCs was done in Norway (J. Skaare).

An interlaboratory calibration study between NSVS and the NWRC laboratories was carried out (C. Sandau, S. Newson, J. Skaare). Both laboratories analyzed aliquots of 21 plasma samples from Svalbard. These were part of the 1998 sample set used for the OH-PCB study in polar bear plasma (see below). Fewer target compounds were included in the Norwegian analysis; however, the number of PCB congeners (15) in common between the two laboratories constituted > 95% of total PCBs.

Hydroxy-PCB metabolites, thyroid hormones and vitamin A

Identification of 22 OH-PCB congeners in polar bear plasma was possible with existing standards. This included all of the major OH-PCBs. OH-PCBs, pentachlorophenol and other chlorinated phenolic compounds were determined in 38 polar bear plasma samples from Svalbard, Norway, and 34 samples from Barrow Strait, Canada (C. Sandau, S. Newson). Polar bears included males and females from 0 to 27 years of age. The chemical analysis also included the quantitation of 24 PCBs, DDT and metabolites, 10 chlordane type compounds, OCS (octachlorostyrene), α-HCH (α-hexachlorocyclohexane), β-HCH, tetrachlorobenzene, pentachlorobenzene and HCB (hexachlorobenzene). Retinol (S. Trudeau), T4, T3, T4 index, and T3 index (S. Brown) were also determined for a majority of the samples.

Prior to the extraction of the above samples, methodology for the efficient extraction and quantitation of the neutral and phenolic chlorinated compounds was optimized using pooled polar bear plasma (C. Sandau, S. Newson). The neutral fraction (PCBs, chlordanes, DDE and chlorobenzenes were quantitated using the previously described Polar Bear Quantitation method (Norstrom et al. 1998). Fully ^{13}C-labelled chlorobenzene and PCB standards were spiked to the plasma samples prior to extraction and were used as internal recovery standards. OH-PCBs and other phenolic compounds were quantitated using authentic standards as described elsewhere (Sandau et al. 2000a). Fully ^{13}C-labelled OH-PCB standards (Wellington Laboratories) were spiked to the plasma samples prior to extraction as internal recovery standards.

During methodological development, an unknown heptachlorinated compound was discovered at relatively high concentrations compared to the identified OH-PCBs. High-resolution mass spectrometry determined the molecular weight from which the identity was hypothesized. The most probable compound was synthesized (Wellington Laboratories); the identity was confirmed by comparing the high-resolution mass spectra and relative retention times on three different gas chromatography columns of varying polarity. The presence of transthyretin (TTR) in polar bear plasma was established by gel electrophoresis of plasma proteins, and the binding affinity of the heptachloro compound to human TTR was determined (I. Meerts and A. Brouwer). Concentrations were determined in polar bear plasma from the Resolute Bay area (M. Ramsay) and compared to ringed seal plasma from the Kuujjuaq area (D. Leclair).

Testosterone in vitro metabolism

The rates of formation of eight different metabolites of testosterone (7α-, 11α-, 16α-, 2β-, 6β-, 16β-hydroxytestosterone, 15β-dihydroxytestosterone and androstenedione) were determined using liver microsomes from 16 male polar bears that had been stored at –75°C to –80°C since 1993 (S. Bandiera, C. Sandau). The separation and quantitation of the metabolites was achieved using high performance liquid chromatography as described previously (Anderson et al. 1998). Each microsomal sample was assayed in duplicate on at least two different days and the results were averaged. Results were compared to previously determined concentrations of CYP1A, CYP2B, a wide range of OCs and EROD activities (Letcher et al. 1996).

PCBs in vitro metabolism

In vitro metabolism of PCBs was investigated using stored liver microsomes from the same 16 male polar bears used for testosterone metabolism (S. Bandiera, C. Sandau). Initial experiments were conducted with pooled rat liver microsomes to optimize assay conditions. Metabolism of several individual PCB congeners was tested by incubating with pooled liver microsomes from polar bears for 60 minutes at 37°C. After extraction and clean-up, the amount of PCB remaining was determined by gas chromatography-electron capture detection (GC-ECD) or gas chromatography-mass spectrometry (GC-MS). The optimization stage is nearly complete using pooled polar bear liver microsomes and the following substrates: CB47, CB52, CB77, α-HCH, β-HCH and *trans*-chlordane. This involves finding the concentration in the medium that is necessary to saturate the enzyme in order to be able to compare rates of metabolism among compounds.

Results

Immunotoxicology

A summary of the NWRC and NSVS interlaboratory comparison of OC and PCB concentrations in 20 plasma samples from Svalbard is shown in Table 1. Lipid concentrations and concentrations of some OCs, especially the HCHs and DDE, were not in good agreement. Excellent agreement was found for HCB, oxychlordane, t-nonachlor, the major PCB congeners and total PCBs. Further work is required to reconcile differences in HCH and DDE concentrations between the laboratories. The discrepancies may be due to the inferior sensitivity of the NWRC method, which employed selected ion monitoring GC-MS; α-HCH and DDE were frequently close to the detection limit of the method. Positive interference in GC-MS analysis may be a problem as well, since the NWRC results tended to be higher (3 × for α-HCH) than those from the NSVS method, which employed GC-ECD. There may also be a discrepancy in standards concentration, which is currently being checked.

The agreement between the two laboratories for PCBs and chlordanes is better than expected, especially since no attempt was made to standardize methodology or

Table 1. Interlaboratory comparison of methods for determination of organochlorine concentrations in duplicate samples of polar bear plasma (n = 20), Svalbard, Norway*

	NWRC**		NSVS***	
	Mean	**SD**	**Mean**	**SD**
Lipid (%)	0.60	0.20	0.96	0.32
HCB	2.14	1.79	2.37	1.95
α-HCH	0.68	1.25	0.22	0.10
β-HCH	2.03	1.47	0.99	0.61
oxychlordane	9.61	7.40	10.46	11.20
t-nonachlor	1.10	0.51	1.14	0.72
p,p'-DDE	0.96	0.55	0.51	0.39
CB99	6.55	4.09	8.07	5.84
CB153	39.48	23.81	38.83	25.27
CB138/163	7.77	4.22	7.06	4.02
CB180	28.63	18.82	28.74	17.42
CB170/190	15.90	11.77	15.37	9.71
CB194	4.97	4.82	7.23	7.54
ΣPCBs	108.42	62.63	110.42	65.88

* No attempts were made to coordinate methods or standards, resulting in a completely unbiased comparison.

** National Wildlife Research Centre

*** Norwegian School of Veterinary Science

Table 2. Pearson correlation coefficients and significance levels between the different organochlorines and G (IgG) in blood plasma of polar bears (n= 56), Svalbard, Norway*

	r	*P*
Chlordanes	–0.22	0.108
DDE	0.08	0.540
HCB	–0.36	0.006
HCHs	–0.17	0.195
PCB-99	–0.40	0.002
PCB-105	–0.13	0.355
PCB-118	–0.04	0.783
PCB-128	–0.10	0.451
PCB-138	–0.26	0.056
PCB-153	–0.26	0.055
PCB-156	–0.26	0.052
PCB-157	–0.06	0.679
PCB-170	–0.25	0.063
PCB-180	–0.25	0.068
PCB-187	–0.11	0.413
PCB-194	–0.28	0.040
PCB-206	–0.28	0.036
PCB-209	–0.18	0.177
ΣPCBs	–0.29	0.029

* IgG was corrected for age and sex by regression (Bernhoft et al. 2000).

use the same standards. Since these classes of chemicals are the main ones found in polar bears, the results of the OC and PCB analyses between the two laboratories can be considered directly comparable, with minor caveats for HCHs and DDE. Because of difficulties in consistency of lipid determinations among laboratories, it is recommended that plasma concentrations always be given on a wet weight basis, and lipid concentration as additional information.

Relationship between immunoglobulin levels and PCB exposure

Immunoglobulins in the blood constitute the combined amount of antibodies against environmental bacteria, viruses and parasites to which the polar bear has been exposed during its life. In young animals, the immunoglobulin concentrations are low but increase rapidly to adult levels within a few years. The ability to produce antibodies is influenced by several factors such as nutritional condition, maternal immunity, hormonal dysfunction, exposure to microbes and parasites, immunosuppression induced by infections, exposure to environmental pollutants and stress conditions. A significant decrease of total IgG in blood plasma of 56 polar bears at Svalbard with increased PCB level has been found (Bernhoft et al. 2000). The PCB congener with highest inverse correlation to IgG was CB99 (Table 2). A similar high inverse correlation between IgG and PCB-99 was found in the present study that used serum samples collected during a 4-year period from 185 polar bears at Svalbard, from 94 polar bears from Lance-cruise 1997-1998 and Svalbard 1998, and from 63 polar bears from Canada 1998-1999. Statistical analysis is not complete at this time for the Canadian bears. Further characterization of these findings will be carried out, correcting for condition, age, sex and some health parameters.

This indicates that the PCB burden in polar bears has a much greater negative effect on the immune system than we had expected. It is, therefore, very important to characterize and document the effects of OCs on the immune system in detail and to study the effects of these changes on the resistance of the Svalbard polar bears to infection.

Relationship between PCB exposure and the ability to produce antibodies

Another part of the investigation deals with the characterization of a possible immunosuppressive effect of PCBs. The experiment is designed to measure specific immune responses in bears from areas with different PCB exposure. It is based on a vaccination model with recapture 6 weeks after immunization and includes polar bears from Canada (1999) with low PCB levels in order to compare them with the Svalbard bears (1998) with high PCB levels. The bears were immunized with different herpes-, reo- and influenza viruses, tetanus toxoid and KLH.

The polar bears responded after 6 weeks by producing protective antibodies, like virus neutralizing antibodies, virus hemagglutinations inhibition antibodies and toxin neutralizing antibodies. In vitro lymphocyte stimulation was also performed with specific antigens. We are now analyzing the PCB content of the blood samples taken at the time of the immunization to see if PCB concentrations correlate with antibody production after immunization, and thereby indicate possible immune suppressive effects in the period up to recapture. By comparing measurements of antibody response against viruses, toxoids and KLH with the results from lymphocyte responses, we will see the effect of PCBs on the combined immune system. In addition, the components of lymphocyte functions that are most affected can be determined. This experiment also includes hematology, hormone analyses and the measurement of Ig levels in the blood. Statistical analysis of these data are underway.

Relationship between PCB exposure and lymphocyte proliferation

The responsive capability of lymphocytes was measured by lymphocyte proliferation testing after in vitro stimulation with different mitogens (PHA, Con A, PWM, PPD and LPS). Although the experiments were carried out under field conditions, the proliferation responses and variation within and between animals was of the same size as in experiments done in human, mice and production animals. PHA-, Con A- and PWM-induced strong lymphocyte response, whereas PPD- and LPS-induced lymphocyte proliferation comprised only smaller populations of cells. The effects of condition and age on the lymphocyte proliferation could be demonstrated. Test results from four field experiments (Lance-cruise 1998, Svalbard 1998, Canada 1999) were included in this investigation of a total number of 121 polar bears. Preliminary analyses of the data show significant lower responses to PPD and LPS in polar bears with high PCB exposure. All lymphocyte data will be corrected for animal condition, sex and age, hormone values, and hematology parameters before the work is completed.

Effect of in vitro exposure of lymphocytes with PCB congeners

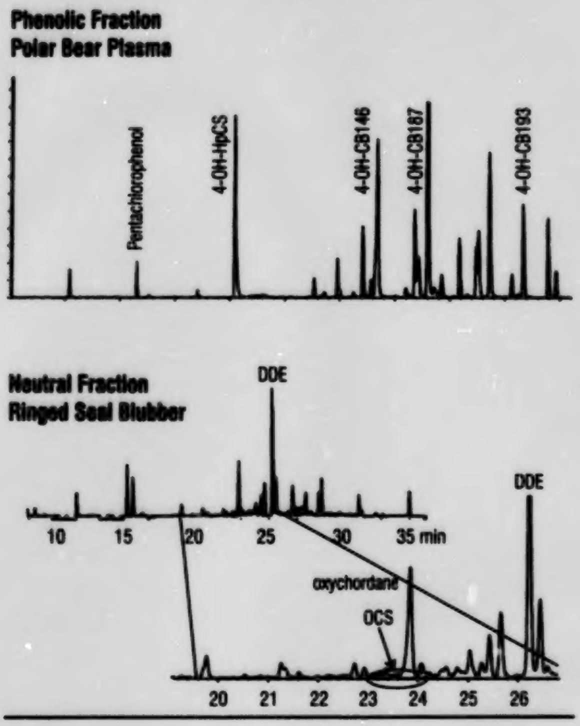
The aim of this study was to investigate the effect on the in vitro PCB exposure on Con A- induced lymphocyte proliferation in polar bears, and to see if the effect on the proliferation response would vary with the in vivo PCB exposure. Test results from three field experiments (Lance-cruise 1998, Svalbard 1998, Canada 1999) involving a total number of 94 polar bears were included in this investigation. Four PCB-congeners were tested and an overall suppression on Con A-induced lymphocyte proliferation could be demonstrated. An effect of the polar bear condition was found so only results from bears that were in good condition were included in the analyses so far. Although the suppression varied, the strongest reduction on the lymphocyte proliferation following in vitro PCB exposure could be demonstrated in animals with high content of PCBs in the blood.

Hydroxy-PCBs

The unidentified phenolic compound was confirmed to be 4-hydroxyheptachlorostyrene (4-OH-HpCS) by synthesis of the authentic compound. Electron impact and electron capture-negative ionization mass spectra and retention times on three columns of widely differing polarity were identical to the compound found in polar bear plasma. The only plausible source of this compound is from metabolism of OCS. The 4-OH-HpCS is a major component of the phenolic fraction of polar bear plasma (Figure 1). The major source of OCS is likely to be older processes of electrolysis of salt solutions using carbon electrodes, primarily in chlorine and caustic soda production, but also in magnesium production. OCS and HCB were produced as a by-product in the viscous substance called taffy-tar which accumulated at the carbon electrode. OCS is commonly determined in environmental samples but seldom reported since it is generally found at low concentrations. The main source of OCS for the polar bear is its diet of ringed seal. In the bottom half of Figure 1 is the chromatogram of the neutral fraction of ringed seal blubber, indicating that OCS is a very minor compound in ringed seal, and therefore in the polar bear diet.

Five ringed seal blood plasma samples from the Kuujjuuaq area of northern Quebec (D. LeClair) were analyzed for OCS and 4-OH-HpCS to compare with plasma concentrations in polar bears (n = 30) from Barrow Strait (Table 3). As seen in Table 3, 4-OH-HpCS is a large proportion of the quantitated phenolic compounds in polar bear plasma and ringed seal plasma, constituting approximately 10% and 16% of the total, respectively. The ratio of 4-OH-HpCS to its precursor OCS is 150-fold higher in the polar bear

Figure 1. GC-MS (ECNI) chromatogram of polar bear plasma (top) and GC-ECD chromatogram of ringed seal blubber (bottom), showing the relative importance of metabolite (4-OH-HpCS) in bear to precursor in diet (OCS)



plasma than in the seal plasma, indicating a higher rate of formation of 4-OH-HpCS in the polar bear.

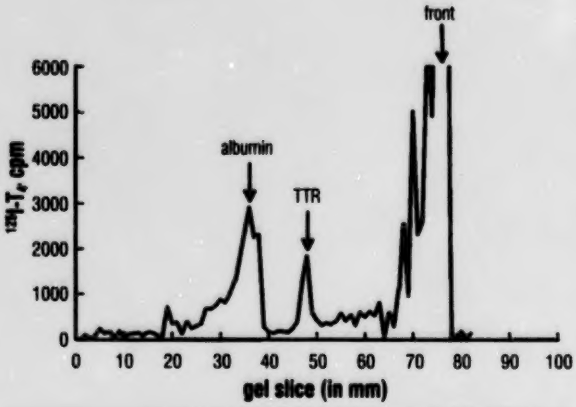
The mechanism of retention of 4-OH-HpCS is likely the same as OH-PCBs. Retention of metabolites is thought to involve competitive binding of the chlorinated phenolic compounds to one of the major thyroid hormone transport proteins, TTR. Polar bear plasma has never been analyzed for thyroid hormone transport proteins, thus six plasma samples were analyzed for the presence of TTR and the other transport proteins. Figure 2 shows the polyacrylamide gel electrophoresis electrophoretogram of one of the plasma samples analyzed after incubation with ^{125}I-labelled thyroxine (T_4), the natural ligand of TTR. As shown in the diagram, two transport proteins were identified in the plasma samples by comparison with authentic standards of human binding proteins. The proteins corresponded to a higher capacity binding protein albumin, and a lower capacity binding protein corresponding to TTR (capacity determined by the intensity of the peak).

The structural requirement for competitive binding with T_4 to TTR is an aromatic OH-group with adjacent halogen atoms. If the compound has two phenyl rings (biphenylether or diphenylether), preferential binding occurs if the OH-group is *para* (4-) to the ring-ring connecting bonds. Since 4-OH-HpCS fits the requirements for binding, competitive binding of 4-OH-HpCS relative to ^{125}I-T_4 for human TTR was determined. 4-OH-HpCS was found to have 1.1 × the binding affinity of T_4 for TTR. Thus, 4-OH-

Table 3. Plasma concentrations (ng·g^{-1} wet weight) and selected mean ratios of concentrations of chlorinated phenolic compounds in polar bear plasma (n = 30), Resolute Bay, Nunavut, and ringed seal plasma (n = 5), Kuujjuaq, Quebec

	Polar Bear Plasma				Ringed Seal Plasma	
	Mean	Minimum	Maximum	SD	Mean	SD
4-OH-HpCS	9.11	2.89	22.9	3.85	0.062	0.023
PCP	0.210	0.093	0.531	0.099	0.237	0.136
ΣOH-PCBs	92.6	26.4	576	117	0.081	0.042
ΣPhenolics	103	33.2	600	120	0.379	0.181
OCS	0.348	0.106	0.940	0.188	0.266	0.086
CB 153	20.2	4.81	82.0	16.3	5.16	3.71
ΣPCBs	46.9	16.1	161	32.3	27.1	16.5
4-OH-HpCS/ΣPhenolics	0.122	0.038	0.248	0.053	0.176	0.077
4-OH-HpCS/OCS	39.2	5.19	216	41.2	0.260	0.124
4-OH-HpCS/CB153	0.712	0.035	2.43	0.580	0.016	0.009
OCS/CB153	0.024	0.005	0.078	0.018	0.072	0.042

Figure 2. Separation by polyacrylamide gel electrophoresis (PAGE) and radioactive detection of proteins in polar bear plasma after incubation with ^{125}I-Thyroxine



HpCS can competitively displace T_4 from the transport protein TTR, which could result in the reduction of circulating thyroid hormone levels in vivo. Low concentrations of lipophilic compounds, such as OCS in polar bear, generally indicate substantial clearance from an organism. However, presence of the probable OCS metabolite in polar bear plasma and its competition with T_4 binding to human TTR demonstrates that biotransformation can result in xenobiotics, which elicit different and potentially more pronounced toxicological activity than their precursor compounds. The discovery, synthesis and binding affinity of 4-OH-HpCS was thoroughly described in a recently submitted publication (Sandau et al. 2000b).

A summary of concentrations of the major OC groups (including OH-PCBs), thyroid hormones and retinol (vitamin A) in Resolute (Barrow Strait) and Svalbard polar bear plasma is given in Table 4. Svalbard bears had significantly higher OH-PCBs then Barrow Strait bears ($p < 0.001$) due to their two- to three-fold higher PCB levels ($p < 0.001$). Juvenile polar bears have similar OH-PCB concentrations as female polar bears even though they have much higher PCB concentrations then both males and females. The juvenile polar bear enzyme system may be immature and not induced to the same extent as adult bears who have incurred a lifetime of exposure to OCs. It is probable that juvenile polar bears are also exposed to OH-PCBs through lactation, since OH-PCBs have been found in milk (Newsome and Davies 1996). The extraction and quantitation of milk samples taken from these same Barrow Strait bears in 1997 may allow us to pursue this theory further.

Female polar bears consistently had higher concentrations of metabolites than males ($p = 0.01$ Barrow Strait, $p = 0.002$ Svalbard) (Table 4). This is opposite to the concentrations

Table 4. Total contaminant classes as wet weight concentrations (ng·g^{-1}) in polar bear plasma, Resolute Bay, Nunavut, and Svalbard, Norway*

	Resolute Polar Bears, 1997						**Svalbard Polar Bears, 1998**					
	Juveniles n = 6		**Males n = 13**		**Females n = 14**		**Juveniles n = 4**		**Males n = 18**		**Females n = 16**	
(ng·g^{-1})	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**
ΣClBzs	7.49	5.15	2.95	0.82	3.19	1.65	4.48	1.56	3.03	2.58	2.05	1.02
ΣHCHs	4.97	2.73	3.59	1.21	2.86	1.15	1.41	0.77	2.71	1.52	1.88	1.91
ΣDDTs	1.48	1.07	0.70	0.40	1.67	3.20	0.82	0.22	1.06	0.60	1.04	0.56
ΣChlordanes	56.23	45.99	13.78	5.79	28.74	16.47	55.36	15.68	19.07	10.64	25.81	8.65
ΣPCBs	79.39	53.79	41.88	15.60	32.75	20.86	228.47	35.57	94.76	49.91	91.55	42.87
ΣOH-PCBs	131.13	154.98	56.84	47.25	112.87	135.17	212.70	184.97	105.47	50.73	217.99	149.66
Ratio OH-PCBs:PCBs	1.62	1.03	1.51	1.23	4.73	5.87	0.85	0.68	1.41	1.11	2.83	2.08
T3 (nmol·L^{-1})	0.17	0.17	0.18	0.18	0.14	0.08	0.23	0.11	0.34	0.30	0.63	0.37
T3 binding index	3.85	1.84	5.06	1.35	2.88	1.46	6.19	0.12	6.31	0.08	6.25	0.09
T4 (nmol·L^{-1})	7.34	2.36	5.02	3.87	6.44	2.02	1.00	1.42	2.33	1.87	3.65	4.74
T4 binding index	1.12	0.09	1.13	0.04	1.15	0.04	0.81	0.10	0.93	0.18	0.83	0.18
Retinol (µg·L^{-1})	354	85	195	59	324	127	302	110	225	98	279	81

* Units of the biological measures are given separately.

of PCBs where males generally have higher PCB concentrations (not statistically significant in this data set, but demonstrated in other analyses [Bernhoft et al. 1996, Norstrom et al. 1998]). Since OH-PCBs are formed by slow metabolism of recalcitrant PCBs, the difference in metabolite formation may be a result of gender-specific metabolism of PCBs where females can metabolize recalcitrant PCBs (e.g. CB-153) more readily than males. Another possible explanation may be that females have a greater binding capacity for OH-PCBs because of higher circulating levels of TTR. Both hypotheses may be pursued in future research and collaboration.

Plasma concentrations of T_3 and the T_3 binding index were significantly higher (both $p < 0.001$) and the T_4 concentrations and T_4 binding index were significantly lower (both $p < 0.001$) in the Svalbard polar bears. The binding indices are measures of residual binding capacity of the plasma for the hormones.

In order to retain all the chemicals in a comparative analysis with the biological measures, principal component analysis (PCA) was used. Since the mechanism of action of the compounds is at a one-to-one molar scale, i.e. one molecule of a metabolite affecting one molecule of T4 binding to TTR, all concentrations of contaminants were changed from mass to molar concentrations. Juveniles were eliminated from the analysis because of their unusually high contaminant levels and possibly unique vitamin A and thyroid hormone stasis. Thus, 60 adult bears from both regions were used in the PCA. The number of samples limits the number of components used in PCA, thus, only 60 contaminants could be used in the analyses. The top 60 compounds were ranked as a molar percentage of the total contaminants quantitated. The top 30 compounds are shown in Table 5.

Table 5. Top 30 compounds found in adult polar bear plasma, Resolute Bay bears (n = 27), Svalbard bears (n = 34)*

Rank	Compound	Mean	SD
1	4-OH-CB187	15.9	12.1
2	CB153	12.3	5.66
3	CB180	7.10	4.13
4	4-OH-CB193	6.75	5.67
5	oxychlordane	5.35	4.02
6	4-OH-HpCS	4.11	2.83
7	CB170/190	3.63	2.77
8	OH-Cl6	3.22	2.30
9	4-OH-CB146	3.12	3.40
10	CB99	2.49	1.09
11	4-OH-CB109	2.34	1.74
12	CB138/163	2.34	0.97
13	4,4'-diOH-CB202	2.14	1.38
14	compound C	2.12	1.48
15	4'-OH-CB199	2.08	1.61
16	4'-OH-CB172	2.07	0.97
17	U4	1.99	2.02
18	β-HCH	1.78	1.43
19	TeCl-benzene	1.18	1.27
20	CB194	1.10	1.04
21	3-OH-CB153	1.00	0.55
22	CB56/60	0.89	2.75
23	hexachlorobenzene	0.88	0.56
24	OH-Cl8	0.81	0.51
25	4'-OH-CB208	0.76	0.72
26	OH-Cl6	0.72	0.48
27	MC-6	0.71	0.43
28	CB52	0.64	1.50
29	3-OH-CB187	0.60	0.47
30	DDE	0.55	0.78

* Compounds are ranked by decreasing molar percentage of total quantitated contaminants.

The PCA yielded four explained (and statistically significant) principal components. Compounds that loaded highly on one principal component (r > 0.70) and low on any other (r < 0.30) were considered significantly and uniquely related to that principle component and are listed in Table 6. Thus, 60 contaminants were reduced to four major principal components, which explained 65% of the total variance of the chemical residue data set. The components are clearly related to chemical class: persistent PCBs, OH-PCBs, non-persistent PCBs (i.e., that have a low biomagnification factor relative to CB153), and chlordanes in decreasing order of explanation of variance. The principal components were then correlated with the biological measures to determine possible relationships.

There was no significant difference in retinol concentrations between regions (Table 4). However, retinol concentrations were found to be significantly inversely correlated ($r = -0.40$, $p = 0.002$) with persistent PCBs (PC1) (Figure 3) and significantly directly correlated ($r = 0.37$, $p = 0.004$) with OH-PCBs (PC2, not shown) for all the bears analyzed for retinol (n = 60). A gender difference in vitamin A concentrations may also be present and statistical analysis is still ongoing. These results are in agreement with the analysis of Skaare et al. (1994), who found a negative correlation between retinol and total PCBs in plasma of polar bears from Svalbard.

Table 6. Principal components analysis of concentrations of the top 60 organochlorine residues in adult polar bear plasma, Resolute Bay bears (n = 27), Svalbard bears (n = 34)*

Principal Component 1		Principal Component 2		Principal Component 3		Principal Component 4	
Persistent PCBs		OH-PCBs		Non-persistent PCBs		Chlordanes	
Component	Coefficient	Component	Coefficient	Component	Coefficient	Component	Coefficient
CB157	0.94	4'-OH-CB172	0.92	CB52	0.85	oxychlordane	0.86
CB170/190	0.94	4'-OH-CB199	0.90	CB56/60	0.81	MC-6	0.86
CB180	0.94	unknown OH-Cl_8	0.83	CB105	0.79	photoheptachlor	0.84
CB156	0.91	4'-OH-CB208	0.82	CB74	0.75	C4	0.71
CB153	0.90	unknown OH-Cl_8	0.81	CB101	0.74	C5	0.70
CB99	0.83	4-OH-CB159	0.79	CB149	0.74		
CB138/163	0.83	4-OH-CB187	0.77	CB118	0.73		
CB194	0.76	4-OH-CB193	0.77	CB47/48	0.73		
CB146	0.72	3-OH-CB153	0.76				
		3-OH-CB187	0.74				
		4-OH-CB109	0.72				
		3'-OH-CB180	0.72				
		4'-OH-CB130	0.71				
% Variance	**28.8**		**15.2**		**11.5**		**9.14**
Eigenvalue	**17.55**		**9.30**		**7.03**		**5.57**

* Variables were given equal weight. Compounds which correlated ($r > 0.70$) with one principal component but not the others ($r < 0.3$) are included. The total variance explained by each principal component is given.

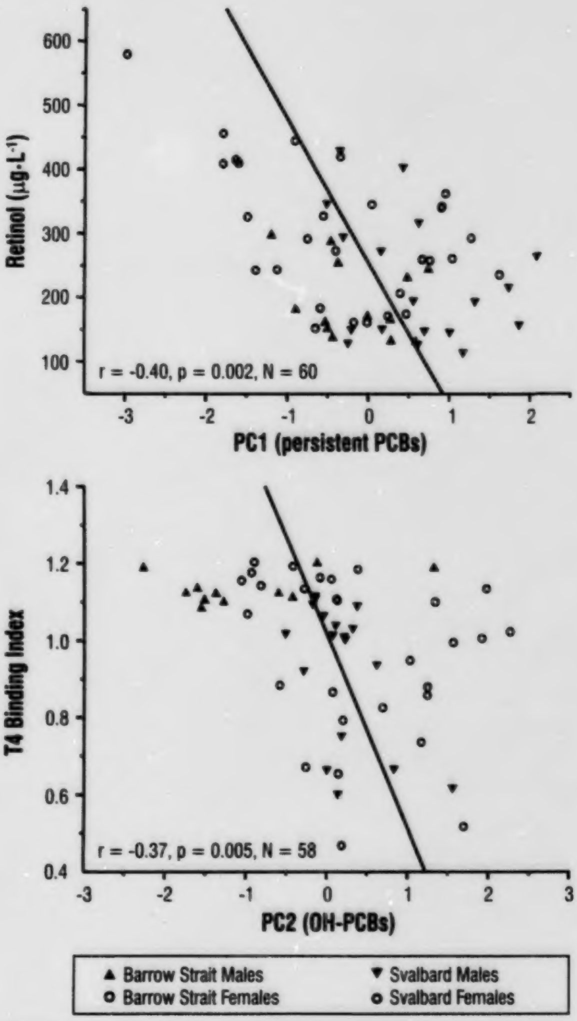
The T_4 index was found to be inversely correlated with PC2 ($r = -0.37$, $p = 0.005$) (Figure 3). The T_4 index is a measure of the binding capacity of the plasma for the hormone, thyroxine. Thus, the negative relationship supports the proposed mechanism of OH-PCBs occupying binding sites in the plasma causing a decrease in the binding capacity of the plasma. The variance of the T_4 index is much higher for the Svalbard samples. This indicates that the index is not a reliable measure of binding capacity in polar bears with higher contaminant levels. The clustering of the Svalbard data below those of the Barrow Strait bears and the lack of correlation between T4 binding index and PC2 in Barrow Strait bears may be an indication of partial deactivation of the binding proteins in some of the Svalbard samples. The Svalbard samples were taken in 1998 for other purposes and not preserved at ultra-low temperatures. Although the Barrow Strait samples were taken in 1997, they were stored at – 40°C or colder from the time of sampling until they were thawed for analysis. Further research is necessary to determine whether this correlation is real or an artifact of sample handling.

Testosterone in vitro metabolism

The major metabolites determined using liver microsomes from polar bear are 2β-hydroxytestosterone, followed by androstenedione and 6β-hydroxytestosterone. Minor metabolites are 7α-, 16α-, 16β- and 11α-hydroxytestosterone. All individual bear microsomal samples were viable with regard to testosterone hydroxylase activities. The activity level was variable among the bears. The results are summarized in Table 7.

In rats, testosterone 2β- and 6β-hydroxylation is catalyzed predominantly by CYP3A enzymes, testosterone 16β-hydroxylation and androstenedione formation is catalyzed by CYP2B enzymes, testosterone 7α-hydroxylation is catalyzed exclusively by CYP2A1, and testosterone 2α-hydroxylation is catalyzed exclusively by CYP2C11. CYP2C11 and CYP2B both catalyze testosterone 16α-hydroxylase activity. The profile of testosterone metabolites produced by polar bears is different from that of male rats (the best studied mammalian model) in which 16α-, 2α-, 6β-, and androstenedione, in that order, are the major metabolites, but is similar to that reported for sheep, goats, and cattle. A recent study with ringed seals showed that 2β- and 6β- hydroxytestosterone were also the major metabolites of in vitro testosterone hydroxylation in this species (androstenedione formation was not monitored).

Figure 3. Inverse correlation of plasma vitamin A concentrations with PC1 (persistent PCBs), and inverse correlation of the plasma T_4 binding index with PC2 (OH-PCBs)



Correlational analysis of testosterone hydroxylase activities with other monooxygenase activities as well as with hepatic OC levels was carried out. No correlation was found between any of the testosterone hydroxylase activities and EROD activity; this is not surprising as none of the testosterone hydroxylase activities are known to be catalyzed by CYP1A enzymes. There was a reasonably good correlation between all of the testosterone hydroxylase activities and BROD (7-benzyloxyresorufin O-deethylase) activity (r values of 0.53 to 0.79), but there was no correlation with PROD (7-pentoxyresorufin O-deethylase) activity, which has previously been shown to correlate with CYP1A (Letcher et al. 1996). There was a weak correlation between testosterone 16α-hydroxylase activity and androstenedione formation with CB 156, CB157, CB170 and CB 180 levels (r values of 0.40 to 0.69). A similar correlation was found between testosterone 2β-, 6β- and 15β-hydroxylase activities with CB 74, CB 85, CB 154, CB 149 and CB 118 (r values of 0.33 to 0.65). A slightly weaker correlation was found between testosterone 2β-, 6β-, and 15β-hydroxylase activities with DDT and DDT metabolites (r values of 0.11 to 0.30). We can conclude from the correlation analysis that i) testosterone 2β-, 6β-, and 15β-hydroxylase activities are catalyzed by different CYP enzymes than testosterone 16α-hydroxylase activity and androstenedione formation in polar bear liver, and that ii) these two groups of CYP enzymes are induced by different PCB congeners. We hypothesize that testosterone 16α-hydroxylase activity and androstenedione formation are catalyzed by CYP2B enzymes, which are induced mostly by di-*ortho* PCBs, and that testosterone 2α-, 6α-, and 15α-hydroxylase activities are catalyzed by CYP3A enzymes, which are induced by PCB congeners that are not considered (with some exceptions) to be CYP1A or CYP2B inducers.

Table 7. Average testosterone hydroxylase activity in microsomes from adult male polar bears (n = 16), Resolute Bay, 1993

	Average activity (nmol metabolite formed·min⁻¹·mg⁻¹ protein)				
Metabolite	**Mean**	**SE**	**Median**	**Minimum**	**Maximum**
15β/DHT	0.23	0.06	0.17	0.06	0.84
6β	4.76	0.56	4.42	1.61	11.23
7α	0.59	0.08	0.54	0.15	1.49
16α	1.53	0.14	1.46	0.74	2.52
16β	0.41	0.04	0.36	0.17	0.76
11α	0.39	0.05	0.35	0.12	0.96
2β	6.13	0.77	5.57	1.82	14.89
Androstenedione	5.35	0.35	5.60	2.76	8.17

PCB in vitro metabolism

The optimization of the metabolism assay has been the main focus of the research of the in vitro metabolism in the last year. It was found that much higher concentrations of PCBs and OCs are needed to saturate the microsomes in order to remain in first order kinetics. The optimization stage is nearly complete using pooled polar bear liver microsomes and the following substrates: CB47, CB52, CB77, α-HCH, β-HCH and *trans*-chlordane. The time of incubations, amount of substrate and concentration of microsomal protein have all been optimized. The next phase will involve the triplicate incubation of the five listed substrates with 14 individual polar bear microsome samples and comparing the average activity for each of the substrates with the contaminant concentrations in the liver and the previously determined cytochrome protein concentrations (1A, 2B) as well as the EROD, PROD and BROD activity. The anticipated completion date is July 2000 with manuscript submission by the end of the fiscal year.

We are finding that the number and position of chlorine atoms on the biphenyl has a great effect on the rate and extent of metabolism. For example, a trichlorobiphenyl such as CB 31 is almost completely metabolized (approximately 90% to 95%) after a 60-minute incubation at initial PCB concentrations ranging from 1 to 35 $ng \cdot mL^{-1}$ (4.25 μM to 136 μM), whereas a hexachlorobiphenyl such as CB 153 is essentially unmetabolized (< 1 %) after a 60 minute incubation at similar PCB concentrations.

We have optimized the assay conditions using pooled polar bear liver microsomes and a series of tetrachlorobiphenyls. Assay conditions were selected and tested to ensure that substrate (PCB) and cofactor (NADPH) concentrations were saturating, and that PCB metabolism was linear with respect to both incubation time and microsomal protein concentration. The final assay conditions for incubation with tetrachloro-PCBs are as follows: i) assay buffer, 0.1 M Hepes Buffer, pH 7.8, with 5 mM $MgCl_2$; ii) final microsomal protein concentration (in the incubation mixture), 0 to 0.5 $mg \cdot mL^{-1}$; iii) length of incubation, 0 to 30 minutes at 37°C with shaking; iv) final substrate (PCB) concentration (in the incubation mixture), 20 to100 μM; v) final cofactor (NADPH) concentration (in the incubation mixture), 1 to 2 mM; and vi) stop solution, 4 mL of hexanes

Discussion and Conclusions

When the data are completely analyzed, the immunotoxicology experiments will contribute to a better understanding of the immune system and hormone function in polar bears with high pollution levels, compared to polar bears with lower pollution levels, and thereby provide insight into the effects of pollution. Norwegian scientists will develop both reagents and methods to measure polar bear immune function, and provide a monitoring method for assessing population status over time. Immuno-pathological studies of PCB effects on the polar bear immune system, immune response ability and resistance to infection are important bases for development of good monitoring methods. Because the Norwegians are the principal investigators in this study, Canadian involvement will be minimal in the future apart from collaboration on preparation of manuscripts. No further studies are planned until all results of the first study are fully interpreted.

A Ph.D. thesis at Carleton University based on the extraction, identification and quantitation of hydroxylated metabolites of PCBs and other halogenated phenolics, and their effects on thyroid hormone and retinol homeostasis in the polar bear, is near completion and will be defended during the Fall of 2000 by Courtney Sandau. One paper is in press and three more papers have been drafted on work based on polar bear plasma (Sandau et al.). The drafted papers will be submitted for publication within the next fiscal year. This project will then be complete. Polar bear blood was shown to have a relatively high concentration of OH-PCBs, as well as a newly found metabolite of OCS. Principle components analysis showed that retinol concentrations in plasma were negatively correlated to total PCBs (but not OH-PCBs). There is some indication that thyroid hormone binding capacity is affected by OH-PCBs (but not PCBs). Further studies are warranted, but none are planned at this time. One fruitful area may be to study the importance of lactational exposure of cubs to OH-PCB metabolites.

Another important area of research which may develop is the effect of OH-PCBs on phase-II metabolism (inactivation and excretion) of hormones. Recent evidence suggests that sulfation and glucuronidation of estradiol, which are the usual biochemical mechanisms of removal, are significantly inhibited by low concentrations of OH-PCBs, which could lead to increased levels of cellular estradiol. A similar mechanism may be operable in excretion of testosterone metabolites, with unknown consequences. Collaborative studies on phase II metabolism of OH-PCB metabolites are under development.

A collaborative study has been initiated with the United States Centers for Disease Control and Prevention in Atlanta, Georgia. Existing extracts (both neutral and acidic phases) from 15 polar bears will be used to test 2D-comprehensive gas chromatography as an emerging technology in the separation and characterization of OCs and their metabolites in plasma. The new technology has never been used on real tissue extracts for the determination of OH-PCBs and other halogenated phenolics. It will allow the determination of the total number of compounds found in polar bear plasma as well as the acquisition of fullscan mass spectra of each of the components once the technology is coupled with time of flight-mass spectrometryResearch will also continue with the identification of the remaining OH-PCBs as well as the identification of the unknown brominated phenolic compounds in plasma with future collaborations with Åke Bergman (Stockholm University). It is unlikely that the NCP will be directly involved in these projects, which are state-of-the-art analytical chemistry research.

However, the results from such studies may lead to identification of new compounds of interest to the Arctic.

The testosterone metabolism study is complete, apart from statistical analysis. Multivariate techniques as well as multiple regression techniques will be applied to determine what effect, if any, PCB exposure on testosterone metabolism rates can be deduced, and which enzymes may be involved. Specific enzymes can be blocked to determine whether they are responsible for production of certain metabolites, but these cannot be connected to exposure to any particular OC. All of the present studies were conducted with samples from adult males. Because field evidence shows that males and females have quite different metabolic capabilities, especially in relation to chlordanes, it would be interesting in future research to repeat this study with females.

It appears that differences in assay conditions will have to be incorporated to measure in vitro PCB metabolism for the various categories of PCB congeners under optimal conditions. A reasonable compromise is to develop one set of assay conditions for each homologue group (i.e. trichlorobiphenyls, tetrachlorobiphenyls, pentachlorobiphenyls and hexachlorobiphenyls). Similarly, a different set of assay conditions needs to be developed for other OCs, such as chlordanes and HCHs. The chorine substitution pattern has a quantitative effect on the rate and extent of biotransformation (metabolism) of the four tetrachlorobiphenyl congeners studied so far. PCB congeners with *ortho* + *para* chlorines on the same ring are metabolized more readily than PCB congeners with a *meta* + *para* chlorine atoms on the same ring, even though both types of congeners have unsubstituted *ortho* + *meta* vicinal positions available for hydroxylation. We expect, although we have not yet determined, that the pattern of metabolites will be different for these types of congeners. The PCB congener and OC metabolism studies are continuing in 2000-2001 with NCP funding. Experiments include determining the rate and extent of metabolism of PCB 47, 52 and 77 with liver microsomes from individual bears, and determining the identity and quantity of OH metabolites (assuming that authentic metabolite standards are available). Assay conditions for a series of trichlorobiphenyls, pentachlorobiphenyls with pooled polar bear liver microsomes will be optimized, and rate and extent of metabolism determined. In addition, hepatic microsomal CYP-metabolism of other OC compounds such as *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, heptachlor, and α-HCH and γ-HCH can be evaluated. The biotransformation studies can also be extended to include liver microsomes from other species, such as ringed seals or beluga whales, as they become available. Whenever relevant, enantio-specific rates of metabolism and metabolite formation will be determined.

Expected Project Completion Date

2001

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Sandau, C.D., S.A. Brown, M. Ramsay, O. Wiig, A. Derocher, J. Duffe and R.J. Norstrom. Implications of hydroxylated metabolites of PCBs (OH-PCBs) and other organochlorine compounds as endocrine disruptors in polar bears.

Sandau, C.D., S. Bandiera and R.J. Norstrom. The optimization and Vmax and Kmax parameters in the metabolism of CB47, CB 52 and CB77 using polar bear liver microsomes.

Sources, Pathways and Levels of Contaminants in Fish from Yukon Waters

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Abstract

Organochlorine contaminant levels in lake trout muscle from Lake Laberge, Yukon, have dropped up to four-fold over the 5-year period from 1993 to 1998. When the lipid-adjusted organochlorine levels in lake trout from Lake Laberge are compared to other Yukon lakes (1998 and 1999 collections only), levels in the Lake Laberge trout do not predominate as they did in the early 1990s. Concentrations of the more lipophilic contaminants such as toxaphene and polychlorinated biphenyls were highest in lake trout from Kusawa Lake (1998-1999) while lake trout from Atlin Lake had the highest concentrations of the less lipophilic organochlorines (hexachlorocyclohexanes and chlorinated benzenes). Organochlorine levels have declined in Lake Laberge, while levels in lakes used for trend monitoring have remained at a consistent level. A corresponding decline in organochlorine contaminant levels was observed in Laberge burbot liver. Toxaphene and ΣDDT lipid-adjusted concentrations decreased in the burbot liver by 3.5- and 4.3-fold, respectively, over the 9-year period from 1990 to 1999. The decline in organochlorine concentrations in trout is most likely linked to the increasing trout population in the lake since the shut down of the commercial and sport fisheries in 1991, and the resulting changes in trout foraging behaviour.

Objectives

- To document the temporal trends of bioaccumulating substances such as heavy metals (mercury, selenium and arsenic), polychlorinated biphenyls (PCBs), DDT, and toxaphene in lake trout muscle and in burbot from selected lakes in the Yukon Territories, so as to determine whether contaminant levels in the fish and thus exposure to people who consume them, are increasing or decreasing with time.

Introduction

With a few exceptions, minimal or no direct temporal trend information on organochlorine contaminants and heavy metals in fish is available for either the Arctic marine or freshwater environments. Much of the temporal trend data that is available is too limited to be scientifically credible because it is based on two or at most three sampling times. In addition, much of this data is confounded by changes in analytical methodology as well as variability in samples due to age and/or size, or dietary and population shifts (Jensen et al. 1997, Arctic Monitoring and Assessment Programme 1998). By comparison, temporal trend data for contaminants in Lake Ontario lake trout (Borgmann and Whittle 1991) and in pike muscle from Storvindeln Sweden (Arctic Monitoring and Assessment Programme 1998) are available over a 15- to 30-year period.

A temporal trend study of contaminants in fish from Lake Laberge is of particular interest because of the unusually high organochlorine contaminant levels found in the lake trout (muscle) and burbot (liver) in 1991 (Kidd et al. 1993, Kidd et al. 1995). As a result of the high historical levels of commercial exploitation, Lake Laberge has a relatively low abundance of lake trout and a high abundance of species such as burbot and sucker compared to other regional lakes. Because of reduced competition and an increased forage base, Lake Laberge lake trout switched to a strict piscivorous diet and as a result grow faster and are much fatter than lake trout from other non-exploited systems such as Kusawa and Fox Lakes (Kidd et al. 1998). In response to the high toxaphene concentrations measured in lake trout, Health Canada issued a public health advisory, recommending that consumption of lake trout flesh be limited and that no livers be eaten. Ultimately, this led to the closure of the commercial and sport fisheries on Lake Laberge.

It has been postulated that if the lake trout stock in Lake Laberge could be rehabilitated and a predator:prey balance restored to a level comparable with other regional lakes, then contaminant levels should decline (DeGraff and Mychasiw 1994). As a result of the advisory and the subsequent closure of the fishery, lake trout abundance in Lake Laberge has almost doubled between 1991 and 1999 (Can-Nic-A-Nick Environmental 2000). This part of our temporal trend studies is being conducted to determine whether the increase in trout abundance has, as predicted, resulted in reduced contaminant concentrations in Lake Laberge fishes.

Activities in 1999-2000

In 1998 and 1999 the Yukon Contaminants Committee sampled lake trout and burbot from Lake Laberge as part of the Lake Recovery Program to re-evaluate the need for the existing health advisory. Samples were also collected from Quiet, Kusawa, Atlin and Coal Lakes for comparison and as part of ongoing temporal trend studies. Organochlorine and metal analysis of these samples, as well as a number of archived samples from the same lakes for retrospective analysis, have now been completed. All samples, with the exception of the 1990 burbot liver samples (Axys Analytical Services Ltd., Sidney BC), were analyzed at the Freshwater Institute in Winnipeg during the 1998-1999 and 1999-2000 fiscal years.

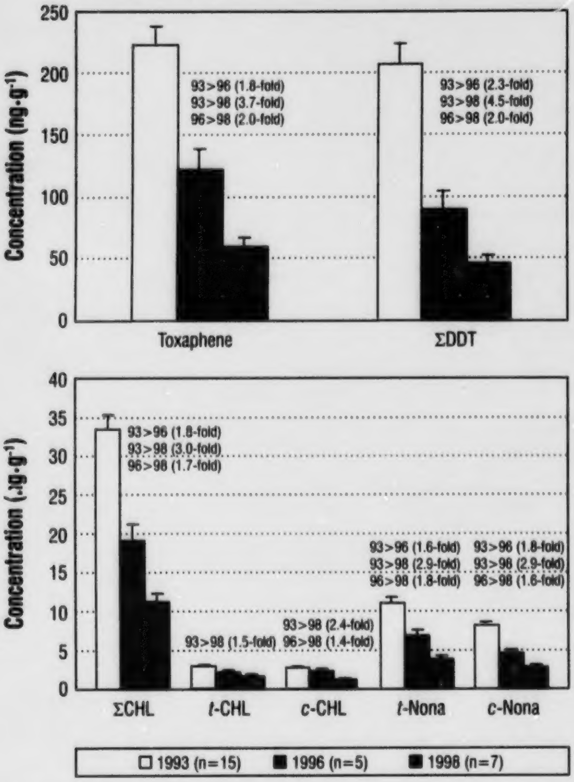
Results

Figures 1 and 2 show the temporal trends of the lipid adjusted least square mean concentrations of selected organochlorines in muscle from Lake Laberge lake trout. Contaminant levels have dropped up to four-fold over the 5-year period from 1993 to 1998. When the lipid-adjusted organochlorine concentrations in lake trout from Lake Laberge are compared with other Yukon lakes (1998 and 1999 collections only), levels in the Lake Laberge trout do not predominate as they did in the early 1990s (Kidd et al. 1993, Kidd et al. 1995). Concentrations of the more lipophilic contaminants such as toxaphene and PCBs were highest in lake trout from Kusawa while lake trout from Atlin had the highest concentrations of the less lipophilic organochlorines (hexachlorocyclohexanes and chlorinated benzenes) (Figure 3). Mean concentrations of major organochlorines and biological data are shown in Table 1. A corresponding decline in organochlorine contaminant levels was observed in Lake Laberge burbot liver. Toxaphene and ΣDDT lipid adjusted concentrations decreased in the burbot liver by 3.5- and 4.3-fold, respectively, over the 9-year period from 1990 to 1999.

Discussion and Conclusions

Organochlorine contaminant levels in Laberge lake trout muscle and burbot liver have decreased significantly since the early 1990s and is most likely associated with the

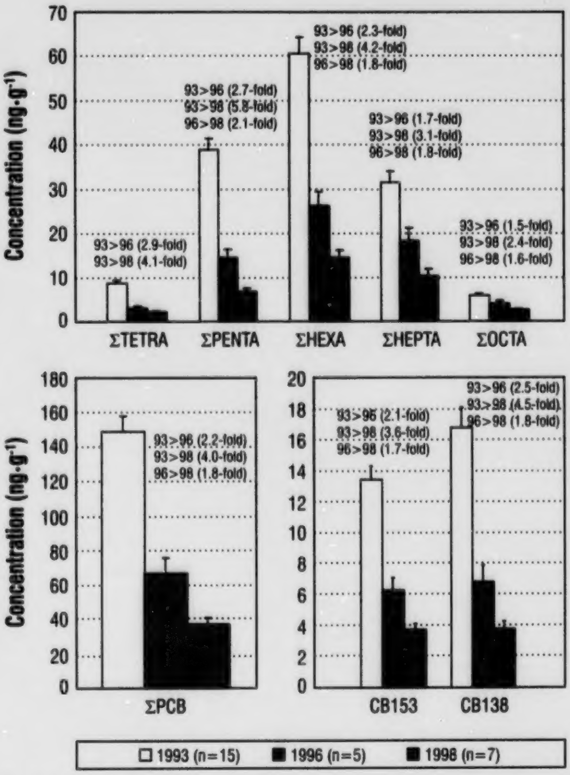
Figure 1. Temporal trends of lipid-adjusted least square mean concentration* of selected organochlorines in lake trout muscle from Lake Laberge**



* Concentration differences are significant by t-test at $p < 0.05$.

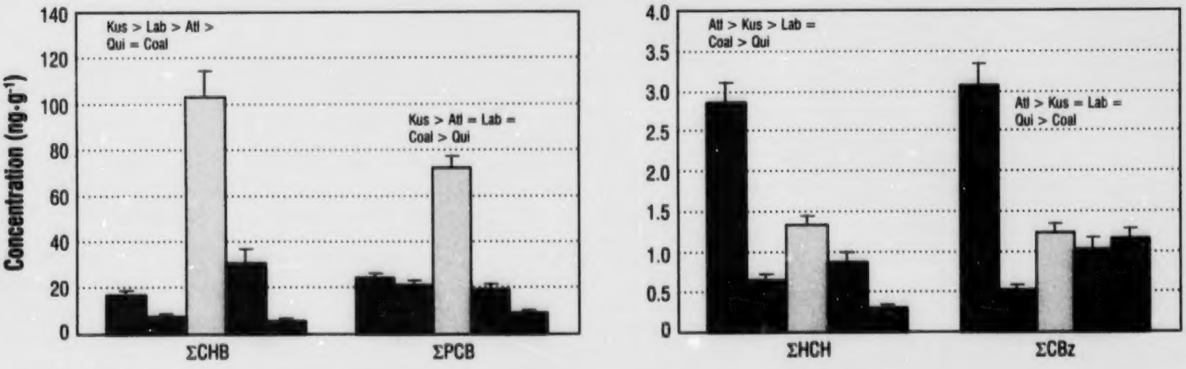
** ΣCHL, chlordanes; *t*-CHL, *trans*-chlordane; *c*-CHL, *cis*-chlordane; *t*-Nona, *trans*-nonachlor; *c*-nona, *cis*-nonachlor.

Figure 2. Temporal trends of lipid-adjusted least square mean concentration* of PCB homologue groups $(Cl_4\text{-}Cl_8)$, ΣPCB and CB 153 and 138 in lake trout muscle from Lake Laberge



* Concentration differences are significant by t-test at $p < 0.05$.

Figure 3a. Spatial trends of lipid-adjusted least square mean concentration* of major organochlorines contaminant groups in lake trout muscle from selected lakes in the Yukon Territory



* LSM concentration differences are significant by t-test at $p < 0.05$.

Figure 3b. Relationship of log [OC] with log % lipid for lake trout in selected Yukon lakes

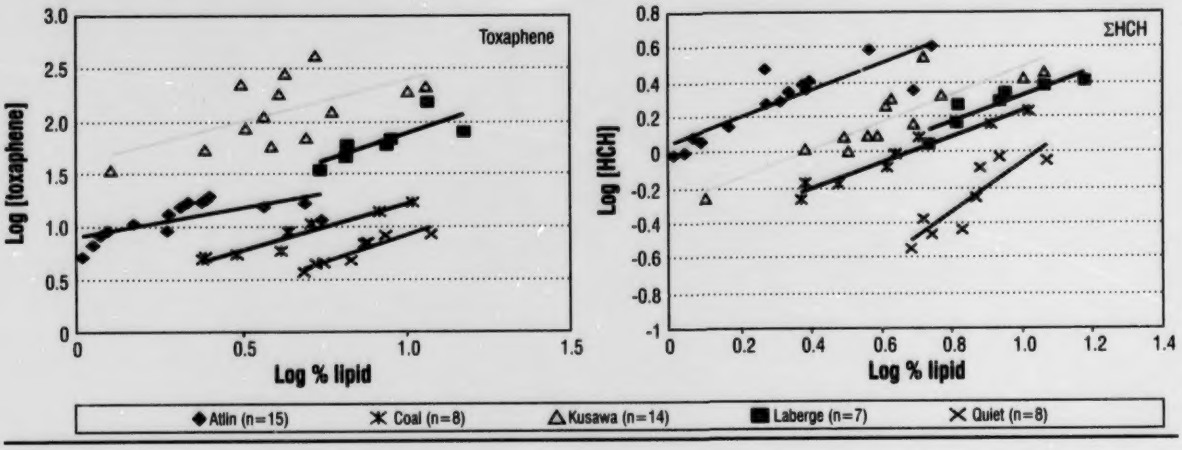


Table 1. Mean concentrations in ng·g^{-1} wet weight (standard deviation) of major organochlorines* in muscle samples from lake trout in the Yukon Territory

Lake	Year	n	% Lipid	Weight (g)	Length (mm)	CF**	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB
Laberge	1993	15	8.22 (4.49)	1563 (1062)	467 (84)	13.22 (2.34)	4.09 (2.77)	5.10 (3.53)	42.88 (27.39)	250.09 (98.41)	182.98 (91.11)	228.73 (66.88)
Laberge	1996	5	8.07 (4.17)	2810 (2122)	554 (123)	14.47 (1.36)	2.24 (0.90)	2.16 (0.82)	22.34 (13.18)	102.13 (51.07)	74.72 (35.12)	125.33 (24.28)
Laberge	1998	7	8.98 (3.37)	4830 (1862)	700 (125)	13.31 (1.30)	2.06 (0.56)	1.92 (0.51)	13.15 (3.62)	52.66 (14.62)	42.58 (10.52)	70.89 (38.22)
Kusawa	1999	14	4.61 (2.98)	1839 (1562)	515 (106)	11.91 (1.51)	1.52 (0.73)	1.68 (0.84)	27.39 (10.25)	139.16 (73.77)	91.09 (44.35)	148.38 (109.59)
Atlin	1998	11	1.76 (0.58)	1940 (2424)	499 (144)	11.96 (1.03)	2.24 (0.84)	1.73 (0.60)	4.71 (2.76)	4.39 (1.58)	17.23 (7.16)	12.67 (4.97)
Quiet	1999	8	2.40 (0.90)	2088 (531)	561 (48)	11.67 (1.17)	0.93 (0.30)	0.25 (0.12)	3.01 (0.72)	1.62 (0.79)	7.24 (2.27)	5.99 (1.88)
Coal	1999	12	3.97 (2.79)	467 (319)	332 (75)	11.56 (2.33)	0.59 (0.44)	0.73 (0.53)	4.67 (5.43)	11.92 (9.70)	25.18 (20.69)	6.78 (4.85)

* ΣCBz, chlorinated benzenes; ΣHCH, hexachlorocyclohexanes; ΣCHL, chlordanes; ΣPCB, polychlorinated biphenyls; ΣCHB, toxaphene.

** Condition factor = [weight/length × 1000 (g·cm^{-3} × 10^3)]

gradual recovery of the lake trout stock since the commercial fishing pressure was lifted. However, this decline in contaminant concentrations is not accompanied by a corresponding decrease in lipid content and condition factor as one might have expected if the trout were reverting back to a diet of increasing amounts of invertebrates.

An in-depth study to examine possible changes in food web structure, lake dynamics, fish populations, and contaminant delivery and loading (air, water, snow, suspended sediments, etc.) is currently underway in an effort to better understand the driving force(s) behind this dramatic decline in organochlorine contaminant levels in Lake Laberge fishes between 1991 and 1999.

Expected Project Completion Date

Temporal trend studies are ongoing and thus this project should continue well into the new millennium.

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Temporal Trends of Polybrominated Biphenyls and Polybrominated and Polychlorinated Diphenyl Ethers in Southeast Baffin Beluga

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Abstract

Blubber samples from male southeast Baffin (Pangnirtung) beluga (1982 [n = 8], 1986 [n = 16], 1992 [n = 11], 1996-1997 [n = 16]) were analyzed for PBDEs (polybrominated diphenyl ethers) and PCDEs (polychlorinated diphenyl ethers). Blubber from five older (> 17 years) male beluga were screened for PBBs (polybrominated diphenyls). ΣPBDE and PBDE47 (2,2',4,4'-TeBDE), the most predominant PBDE congener residue in the beluga blubber, increased by 6.8- and 6.5-fold, respectively, over the 15-year period from 1982 to 1997; this increase reflects the increase in PBDE industrial usage world wide. Conversely, PCDE concentrations in southeast Baffin beluga have shown a significant decline over the same time period. This decrease in PCDE levels reflects the fact that the usage of chlorophenols, which contain PCDE as impurities, has been banned in Canada and Scandinavia, and chlorophenols are now registered only for restricted use in the United States and western Europe. The presence of these contaminant groups in the beluga provide additional evidence in support of their atmospheric transport to the Arctic. PBB levels were found to be below instrument detection limits in all samples.

Objectives

1. To document the temporal trends of PBBs (polybrominated diphenyls), PBDEs (polybrominated diphenyl ethers) and PCDEs (polychlorinated diphenyl ethers) in Arctic marine ecosystems so as to determine whether contaminant levels in marine mammal tissues, and thus exposure of people living in Arctic communities who consume them as part of their traditional diet, are increasing or decreasing with time.
2. To provide data on contaminant levels in marine mammal tissues as part of surveys of dietary contamination and for use by the Arctic Monitoring and Assessment Programme.

3. To equip Canadian delegations involved in international controls with appropriate information to formulate Canadian positions.

Introduction

Marine mammals are harvested by several Inuit communities. These animals occupy high trophic levels in marine food webs and so accumulate (relatively) high concentrations of persistent organohalogens. Documentation of temporal trends in the extent of contamination by these compounds will contribute to an exposure risk assessment.

PBBs and PBDEs

Brominated flame retardants are used in relatively high concentrations in electronic equipment such as computers and television sets, building materials, carpets, cars, aircraft and in many other applications. PBBs and PBDEs are used as additive flame retardants (Sellström 1996), which are incorporated into the matrix of various plastic materials such as polystyrenes (Zitko 1993). Over the last two decades, the use of brominated flame retardants has increased substantially. In Japan, the annual consumption of brominated organic compounds increased from 2500 tonnes in 1975 to 22 100 tonnes in 1987 (Watanabe and Tatssukawa 1989). From 1984 to 1992, the total consumption of PBDEs worldwide doubled from approximately 20 000 to 40 000 tonnes per year. Presently, the worldwide consumption of brominated flame retardant is estimated to be 150 000 tonnes per year (Sellström and Jansson 1995, Sellström 1996, Jansson et al. 1993, Zitko 1993). As a result, PBBs and PBDEs have been released into the environment to such an extent that they are now ubiquitous environmental contaminants.

PBBs and PBDEs are similar in behaviour and toxicity to well-known environmental contaminants such as PCBs (polychlorinated biphenyls) and DDT but to date, have not been banned. Because of their high lipophilicity (log $K_{ow} > 5$, where K_{ow} is the octanol-water partition coefficient) and resistance to degradative process, both groups of chemicals are expected to bioaccumulate easily (Pijnenburg et al. 1995, de Boer et al. 1998, de Boer et al. 2000). According to Safe (1992), the relative toxicities, toxic equivalent factors (TEFs), for the PBBs are similar to those of PCBs. PBDEs and PBBs have been detected at the parts per billion (ppb) levels (lipid wt.) in fish, fish eating birds and marine mammals from remote areas (Pijnenburg et al. 1995, Jansson et al. 1987, Watanabe et al. 1987, de Boer 1989, de Boer et al. 1998, de Boer et al. 2000, Kuehl et al. 1991, Sellström et al. 1993, Loganathan et al. 1995, Haglund et al. 1997), but limited information is available in Arctic marine mammals.

Chlorinated diphenyl ethers

Chlorinated diphenyl ethers are a group of potentially significant organochlorine contaminants. Desired applications of PCDEs have included use as hydrolic fluids, electrical insulators, flame retardents, lubricants, plasticizers and as a pesticide (Koistinen 2000). PCDEs can also be formed in combustion processes (Paasivirta et al. 1986). However, PCDE impurities in technical chlorophenyl formulations are the major source of PCDEs to the environment (Koistinen 2000). Individual PCDE congeners have been reported to be immunotoxic and induce hepatic microsomal enzymes (Howie et al. 1990). Toxicity and induction depends on structure, but relationships are not similar to PCBs. The mono-ortho chloro-substituted PCDE congeners have been shown to be more potent inducers of AHH (aryl hydrocarbon hydroxylase) and EROD (7-ethoxyresorufin o-deethylase) activities and more immunotoxic than their non-ortho analogues (Howie et al. 1990), whereas the non-ortho PCBs are more active than the mono-ortho PCBs. According to Safe (1992), the relative toxicities, TEFs, for the non- and mono-ortho PCDEs are similar to the mono-ortho substituted PCBs ($TEF_{PCDEs} = 0.001$). PCDEs are known to be persistent in the environment and can act as precursors of PCDD and PCDF in photolytic reactions (Humppi and Keinola 1985). As with PBDEs and PBBs, PCDEs have been detected at the ppb levels (lipid wt.) in fish, fish-eating birds and marine mammals from remote areas (Paasivirta et al. 1986, Koistinen et al. 1993, Koistinen et al. 1995, Koistinen 2000), but information available regarding Arctic marine mammals is extremely limited.

Activities in 1999-2000

Archived blubber samples from male southeast Baffin (Pangnirtung) beluga (1982 [n = 8], 1986 [n=16], 1992 [n=11], 1996-1997 [n=16]), previously analyzed for other persistent organic pollutants (POPs) such as ΣDDT, ΣPCBs chlorinated-*n*-alkanes and coplanar PCBs in the 1997-1998 and 1998-1999 fiscal years, were extracted and analyzed for PBDEs and PCDEs. Blubber from selected animals were screened for PBBs.

Collections

Blubber samples from beluga in the Clearwater Fjord area of Cumberland Sound (1982 and 1986) were obtained by Department of Fisheries and Oceans (DFO) personnel (Winnipeg, MB) with the cooperation of local hunters. As part of an ongoing whale sampling and stock identity program, supported by the Nunavut Wildlife Management Board and DFO, samples from the southern coastline in Cumberland Sound (1992, 1996 and 1997) were collected by hunters during their subsistence hunts using standardized whale kits. Blubber, kidney, liver, ovaries and uterus, muscle and the lower jaw, as well as morphometric data were collected for each animal. All samples were shipped frozen to the Freshwater Institute and stored at –40°C until analysis. Animals were aged by thin sectioning a canine tooth from the rear of the lower jaw and counting growth layer groups in the dentine using transmitted light (Heide-Jørgensen et al. 1994).

Analysis

Blubber samples were partially thawed, and 2 grams were combined with anhydrous Na_2SO_4 (heated at 600°C for 16 hours before use). The mixture was then extracted twice with hexane in a small (50 mL) ball mill, centrifuging and decanting the hexane between extractions. Extractable lipids were determined gravimetrically on a fraction (1/10) of the extract. Three hundred mg of each sample extract was sent to the Institute of Ocean Sciences for PBDE and PCDE analysis. Samples were spiked with a suite of internal standards and processed for GC-HRMS (gas chromatography-high resolution mass spectrometry) analysis (Ikonomou et al. 1999). Quantitation was based on custom standard solutions prepared by Cambridge Isotope Laboratories.

Statistical analysis

All univariate analyses were performed with lipid normalized $\log_{10}$ transformed data to adjust for skewness. Analysis of covariance (ANCOVA) was used to assess the effects of year-to-year collections (temporal trends), age of the animals and age*year interactions (homogeneity of slope between age and [organohalogen]) using the model [organohalogen] = year age age*year, where [organohalogen] = log concentration of individual PBDE and PCDE congeners and homologue groups. Differences between collection years were examined with paired comparisons of age adjusted least squared mean concentrations (SAS Institute 1989-1996). Only results for animals older than 2 years of age were included in the analysis of covariance because of the large variations in concentrations seen in younger animals (Stern et al. 1994).

Results

PBDEs

Temporal trends of lipid normalized (age adjusted) PBDE homologue group and congener concentrations are shown in Figure 1. Since 1982, the levels of the major PBDE homologue groups and congeners in the southeast Baffin beluga have increased significantly. Concentrations of PBDE47 (2,2',4,4'-TeBDE), the most predominant PBDE congener residue in the beluga blubber, increase 6.5-fold over this 15-year period (Table 1). These results almost certainly reflect the increase in industrial usage of PBDEs worldwide. Figure 2 shows the percent contribution of the major PBDE congeners and homologue groups to total PBDE in the beluga for each time point (age adjusted). Over the 15-year time span from 1982 to 1997, contributions of the TrBDE homologue group and PBDE47 to total PBDEs have declined by 7% and 3%, respectively. Conversely, PeBDE and HxBDE contributions have increased by 20% and 80%, respectively. This change in the southeaset

Figure 1. Temporal trends of lipid normalized (age adjusted least square mean) concentrations of PBDE homologue groups and major congeners in male southeast Baffin beluga blubber samples

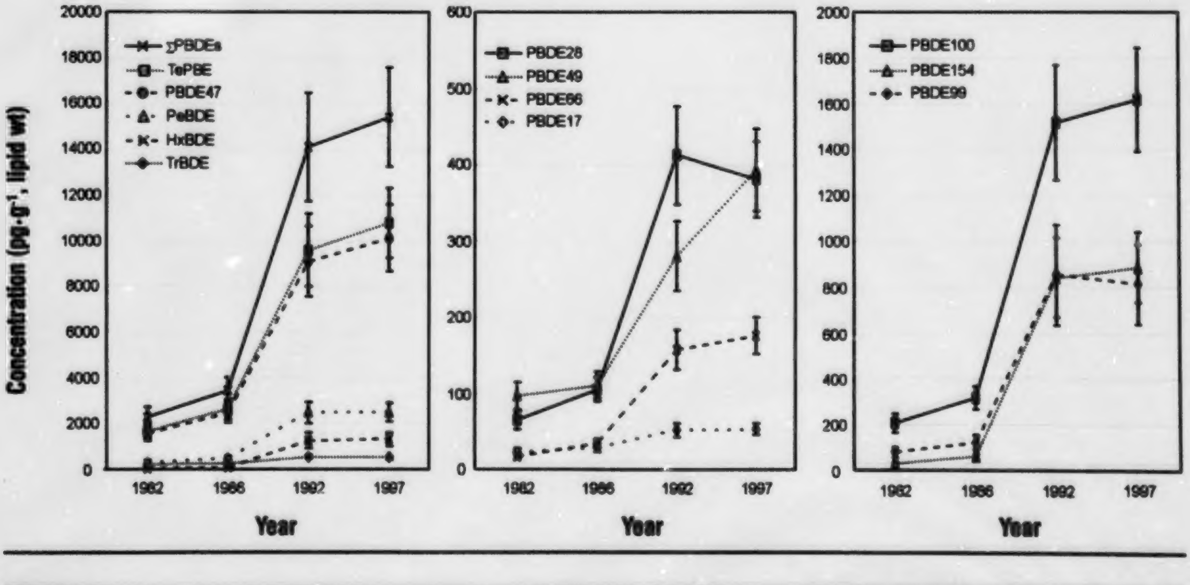


Table 1. Factor increases of major PBDE congeners and homologues between time points*

	PBDE Congeners								PBDE Homologues				
Time	17	28/33	47	49	66	99	100	154	Tr	Te	Pe	Hx	ΣPBDE
1982-1986	1.3	*1.6*	*1.6*	1.2	*2.0*	1.6	1.5	*2.1*	1.1	*1.6*	1.5	*3.0*	1.5
1982-1992	*2.3*	*6.4*	*5.9*	*2.9*	*9.3*	*10.8*	*7.4*	*29.0*	*2.4*	*5.8*	*7.9*	*40.7*	*6.2*
1982-1997	*2.3*	*5.9*	*6.5*	*4.1*	*10.3*	*10.3*	*7.9*	*30.6*	*2.3*	*6.5*	*8.0*	*42.2*	*6.8*
1986-1992	*1.8*	*3.9*	*3.7*	*2.5*	*4.6*	*6.9*	*4.8*	*14.1*	*2.1*	*3.7*	*5.2*	*13.6*	*4.1*
1986-1997	*1.8*	*3.6*	*4.1*	*2.5*	*5.1*	*6.6*	*5.1*	*14.9*	*2.0*	*4.1*	*5.3*	*14.1*	*4.5*
1992-1997	1.0	0.9	1.1	1.4	1.1	0.9	1.1	1.1	0.9	1.1	1.0	1.0	1.1

* Italicized numbers indicate that least square mean concentration differences were significant by t-test at $p < 0.05$.

Baffin beluga PBDE composition is most likely driven by the shift in composition of commercial PBDEs to higher brominated mixtures (de Boer et al. 2000).

PCDEs

PCDE composition (age adjusted) in the blubber samples for each of the four time points are shown in Figure 3. HxCDEs were most predominant followed by the Hp- and PeCDEs. The two most abundant congeners were PCDE 99 (2,2',4,4',5-PeCDE) and PCDE 154 (2,2',4,4',5,6'-HxCDE). This profile is very similar to the PCDE profile observed in the Finnish chlorophenol-based wood preservative, Ky-5 (Koistinen 2000). Over the 15-year period from 1982 to 1997 there has been an 8% increase in the proportion of HxCDEs (other than PCDE 154).

Temporal trends of lipid normalized (age adjusted) PBDE homologue group and selected congener concentrations are shown in Figure 4. Since 1982, the levels of PCDEs in the southeast Baffin beluga have decreased significantly. Concentrations of PCDE99 and PCDE154 have decreased 2.5- and 1.8-fold over this 15-year period (Table 2). This downturn in PBDE levels most likely reflects the fact that the usage of chlorophenols has been banned in Canada and Scandinavia, and are now only registered only for restricted use in the United States and western Europe.

PBBs

Blubber samples from five older (> 17 years) male beluga were analyzed for PBBs. In all samples PBBs levels were found to be below instrument detection limits.

Figure 2. PBDE composition in male southeast Baffin beluga blubber samples collected in 1982, 1986, 1992 and 1997 (age adjusted)

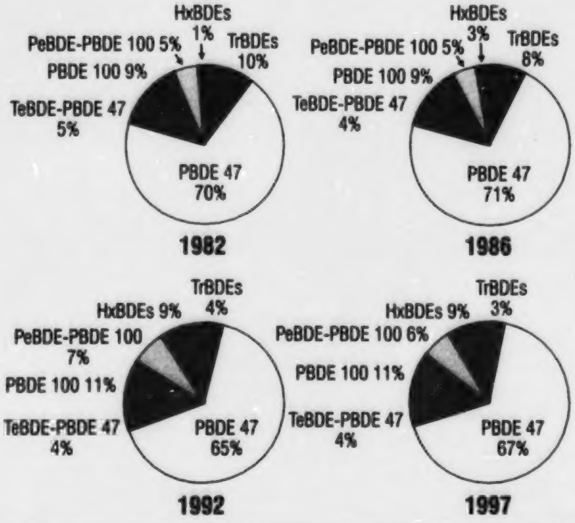


Figure 3. PCDE composition in male southeast Baffin beluga blubber samples collected in 1982, 1986, 1992 and 1997 (age adjusted)

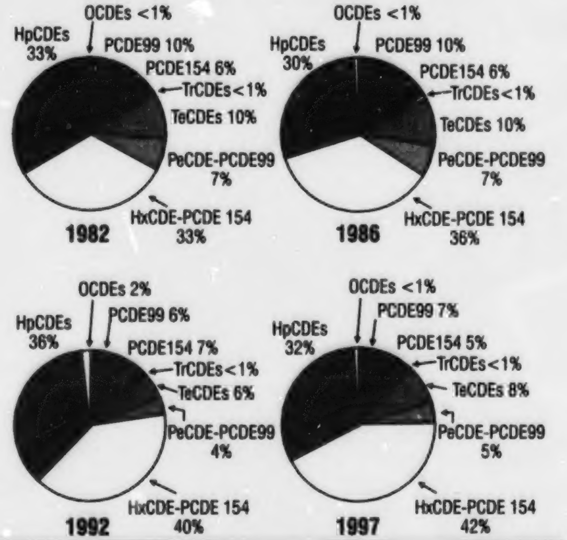
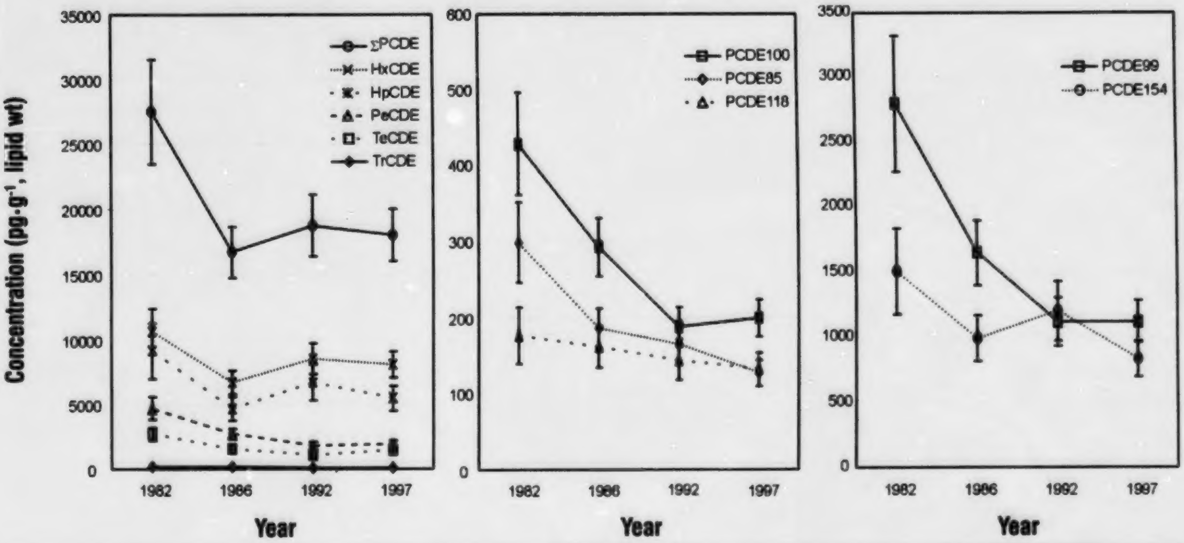


Table 2. Factor increases of major PBDE congeners and homologues between time points*

	PBDE Congeners					PBDE Homologues					
Time	**85**	**99**	**100**	**118**	**154**	**Tr**	**Te**	**Pe**	**Hx**	**Hp**	**ΣPBDE**
1982-1986	*1.6*	*1.7*	*1.5*	1.1	1.5	1.1	*1.7*	*1.7*	*1.6*	*1.9*	*1.7*
1982-1992	*1.8*	*2.5*	*2.3*	1.2	1.3	1.6	*2.5*	*2.6*	1.2	1.4	*1.5*
1982-1997	*2.4*	*2.5*	*2.2*	1,3	*1.8*	1.5	*1.9*	*2.4*	1.3	1.7	*1.5*
1986-1992	1.1	1.5	*1.6*	1.1	0.8	1.5	1.5	1.5	0.8	0.7	0.9
1986-1997	1.5	1.5	*1.5*	1.2	1.2	1.4	1.1	1.4	0.8	0.9	0.9
1992-1997	1.3	1.0	1.0	1.1	1.4	0.9	0.8	0.9	1.1	1.2	1.0

* Italicized numbers indicate that least square mean concentration differences were significant by t-test at $p < 0.05$.

Figure 4. Temporal trends of lipid normalized (age adjusted least square mean) concentrations of PCDE homologue groups and major congeners in male southeast Baffin beluga blubber samples



Discussion and Conclusions

This report provides the first temporal trend data on PBDEs and PCDEs in Arctic marine mammals. Their presence in the beluga also provides further evidence in support of their atmospheric transport to the Arctic. In contrast to PCDEs and other organochlorine compounds such as coplanar PCBs and HCHs (Stern and Addision 1999), the concentrations of PBDEs in eastern Arctic beluga have increased continuously over the 15-year period from 1982 to 1997. The increasing trend is of great concern and calls for measures to stop the usage of PBDEs.

Future work will involve increasing this time series to include one additional collection in 2001. This will give us five time points covering a period of 19 years. In 2000-2001, this study will be expanded to include western Arctic beluga from Kugmallit Bay (1989, 1995 and 2000).

Expected Project Completion Date

Temporal trend studies are long-term propositions and thus this project should continue well into the new millennium.

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Temporal Trends of Organochlorine, Organobromine and Heavy Metal Contaminants in Burbot from Fort Good Hope, Northwest Territories

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Abstract

Muscle and liver samples from burbot collected at Fort Good Hope (Rampart Rapids) in 1999, and a number of archived liver samples from 1988, were analyzed for mercury and selenium. Data from these time points combined with existing data from 1985, 1993 and 1995 cover a time span of 14 years. No temporal trend of mercury was evident in the data from either the muscle or liver. Mean mercury concentrations in muscle and liver over the entire data sets were 0.247 ± 0.073 ($n = 67$) and 0.044 ± 0.019 ($n = 58$) $\mu g \cdot g^{-1}$, respectively. Muscle mercury levels are below the recommended guideline level of 0.50 $\mu g \cdot g^{-1}$ for commercial sale but are at, or exceed, the guideline level of 0.2 $\mu g \cdot g^{-1}$ recommended for fish used for subsistence. Selenium concentrations decreased 1.4- and 1.8-fold between 1985 and 1999, in muscle and liver, respectively. Selenium concentrations in liver declined with increasing length.

Objectives

To document the temporal trends of bioaccumulating substances such as heavy metals (mercury [Hg], selenium [Se], arsenic [As]), PCBs (polychlorinated biphenyls), DDT, toxaphene, coplanar PCBs and selected current use chemicals such as short chain PCAs (polychlorinated-*n*-alkanes), PBBs (polybrominated biphenyls), BDPEs (brominated diphenyl ethers) and CDPEs (chlorinated diphenyl ethers) in burbot liver (and muscle for metals) from Fort Good Hope,. This is to determine whether contaminant levels in the fish, and thus exposure of people living in Arctic communities who consume them as part of their traditional diet, are increasing or decreasing with time. These results will also help to test the effectiveness of international controls.

Introduction

With a few exceptions, minimal or no direct temporal trend information on organochlorine contaminants and heavy metals in fish are available in either the Arctic marine or freshwater environments. Much of the temporal trend data that is available are too limited to be scientifically credible because they are based on two, or at most three, sampling times. In addition, much of this is confounded by changes in analytical methodology as well as variability due to age/size, or dietary and population shifts (Jensen et al. 1997, Arctic Monitoring and Assessment Programme 1998). By comparison, temporal trend data for contaminants in Lake Ontario lake trout (Borgmann and Whittle 1991) and in pike muscle from Storvindeln Sweden (Arctic Monitoring and Assessment Programme 1998) are available over a 15- to 30-year period.

Very few measurements have been made of the toxic coplanar (or non-*ortho* substituted) PCBs and other planar organochlorines in Arctic fish; however, where coplanar PCBs have been measured along with PCDDs (polychlorinated dibenzo-*p*-dioxins), PCDFs (polychlorinated dibenzofurans), (PCNs) polychlorinated naphthalenes and CDPEs, calculations of TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin) equivalents (TEQs) show that the coplanar PCBs, especially CB126, account for most of the TEQs (Jensen et al. 1997). As a result, one of the priorities outlined in both Jensen et al. (1997) and Arctic Monitoring and Assessment Programme (1998) is for additional measurements of coplanar PCBs in Arctic fish. Measurements of PCDDs and PCDFs were not recommended unless specific sources are suspected. Because of their toxicological significance, analysis of new chemical contaminants (i.e. those not currently identified in the United Nations Economic Commission for Europe Convention on Long-range Transboundary Air Pollution protocols) such as PCAs, PBBs, CDPEs and BDPEs are essential and will provide a "baseline" against which future measurements can be compared.

Burbot is a predatory, bottom-feeding species of fish. Its sedentary nature and high lipid content of the liver make the species very suitable for monitoring lipophilic contaminants (Jensen et al. 1997, Arctic Monitoring and Assessment Programme 1998). As outlined in the Northern Contaminants Program Phase II, Call for Proposals (Blueprint for International Activities, and Blueprint for Monitoring the Health of Arctic Peoples and Ecosystems; Northern Contaminants Program 1999), some of the most comprehensive temporal trend data that exists is for organochlorines in burbot liver from Fort Good Hope (1986, 1989 and 1994). Data also exist for Hg and Se in both burbot muscle and liver for various years over the period from 1985 to 1995 (see Tables 1 and 2) (Wagemann 1985, Lockhart et al. 1989, Lockhart 1999).

Activities in 1999-2000

In December 1999, 42 burbot were collected from the Mackenzie River at Fort Good Hope (Rampart Rapids) by community residents. For various reasons, the burbot were not shipped to the Freshwater Institute until late January 2000. As a result, the As and organic contaminant analyses have not yet been completed. Hg and Se analyses have been completed, the results of which are reported below.

Table 1. Mean (standard deviation) Hg concentrations in burbot muscle and liver, Fort Good Hope

Collection	n	Length (mm)	Muscle Hg ($\mu g \cdot g^{-1}$)	Liver Hg ($\mu g \cdot g^{-1}$)	Intercept	Slope	r^2
Apr. 1985*	14	646 (93)	0.233 (0.054) a	—	0.0884	0.0002	0.1516**
Dec. 1993	10	718 (126)	0.251 (0.099) a	—	0.102	0.0002	0.0695
Sept. 1995	4	—	0.223 (0.072) a	—	—	—	—
Dec. 1999	39	708 (107)	0.254 (0.075) a	—	0.1635	0.0001	0.0335
Apr. 1985*	13	654 (92)	—	0.042 (0.018) a	0.0316	2×10^{-5}	0.0062
Dec. 1988	7	726 (65)	—	0.051 (0.021) a	–0.0667	0.0002	0.2646**
Dec. 1999	38	710 (95)	—	0.044 (0.019) a	0.0522	-1×10^{-5}	0.0034

* Source: Wagemann 1985

** Significant at $p < 0.05$

Data in the same column followed by the same letter do not differ significantly by t-test ($p > 0.05$).

Table 2. Mean (standard deviation) Se concentrations in burbot muscle and liver, Fort Good Hope

Collection	n	Length (mm)	Muscle Hg ($\mu g \cdot g^{-1}$)	Liver Hg ($\mu g \cdot g^{-1}$)	Intercept	Slope	r^2
Apr. 1985*	16	663 (111)	0.404 (0.116) a	—	0.5668	–0.0002	0.055
Dec. 1999	42	705 (107)	0.312 (0.137) b	—	0.5766	–0.0004	0.087
Apr. 1995*	15	672 (110)	—	1.564 (0.649) a	3.0007	–0.0021	0.131**
Dec. 1988	10	689 (87)	—	1.276 (0.713) ab	3.3728	–0.003	0.137**
Dec. 1999	39	709 (96)	—	0.879 (0.615) b	2.4688	–0.0022	0.122**

* Source: Wagemann 1985

** Significant at $p < 0.05$

Data in the same column followed by a different letter differ significantly by t-test ($p < 0.05$).

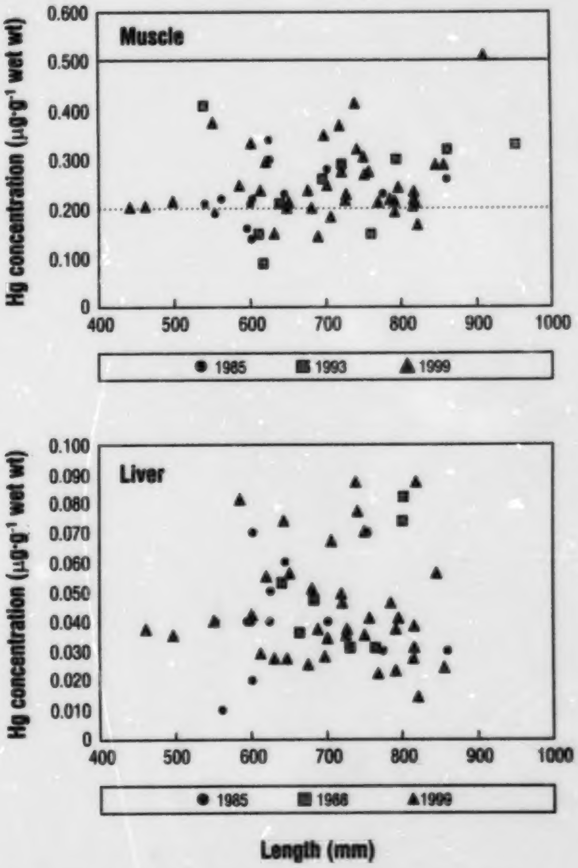
Results

Mean Hg and Se concentrations and regression data for burbot samples collected between 1985 and 1999 are shown in Table 1 and 2, respectively. Hg concentrations are also shown graphically as scatter plots with muscle and liver levels plotted against length (Figure 1). Weak positive correlations exist for the 1985 muscle and 1998 liver samples. No significant differences in Hg for muscle or liver concentrations were observed between time points over the 14-year period from 1985 to 1999 (t-test, $p > 0.05$ in all cases). For Se, weak negative correlations exist between concentration and length for liver samples only. Mean concentrations of Se in both muscle and liver were significantly lower in the 1999 samples compared to those collected in 1985 ($p < 0.05$). Length adjusted least square mean Se liver concentrations (ANCOVA) were 1.336, 1.075 and 0.750 $\mu g \cdot g^{-1}$ for the 1985, 1988 and 1999 time points, respectively. The observed decline in concentrations between 1985 and 1999 was still found to be significant (t-test, $p < 0.05$).

Discussion and Conclusions

Muscle and liver samples from burbot collected at Fort Good Hope (Rampart Rapids) in 1999 and a number of archived liver samples from 1988 were analyzed for Hg and Se. Data from these time points combined with the existing data from 1985, 1993 and 1995 cover a time span of 14 years. No temporal trend in Hg was evident in the data from either the muscle or liver. Mean Hg concentrations in muscle and liver over the entire data sets were 0.247 ± 0.073 (n = 67) and 0.044 ± 0.019 (n = 58) $\mu g \cdot g^{-1}$, respectively. Se concentrations decreased 1.4- and 1.8-fold between 1985 and 1999, in the muscle and liver, respectively. Liver concentrations of Se declined with increasing fish length. A similar correlation has been observed for Atlantic cod (Stange et al. 1996). As was observed in fish muscle from other lakes in the

Figure 1. Muscle and liver mercury concentrations in burbot of varying length collected in Fort Good Hope (Rampart Rapids) at three time points over a 14-year period*



* The solid line at 0.5 $\mu g \cdot g^{-1}$ mercury is the maximum recommended in fish for commercial sales and the dashed line at 0.2 $\mu g \cdot g^{-1}$ is the maximum recommended in fish for subsistance.

Northwest Territories (Lockhart and Evans 1999), no statistical correlation was observed between Hg and Se.

Organohalogen analysis in burbot liver and As analysis in muscle and liver are currently underway and will be reported in 2001. Additional burbot collections from Fort Good Hope (Rampart Rapids) are planned for December 2000, 2001 and 2002.

Expected Project Completion Date

Temporal trend studies are long-term propositions and thus this project should continue well into the new millennium.

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Contaminants in Arctic Seaducks

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Abstract

We examined mercury, selenium and cadmium concentrations and several aspects of health and fitness in common eiders, a seaduck species whose populations have been declining dramatically in the Arctic in recent years. The study was conducted at East Bay Migratory Bird Sanctuary on Southampton Island, Nunavut, in June and early July, 1999. Cadmium concentrations were quite high in these birds compared to conspecifics from other locations. Selenium was lower than in spectacled eiders from western Alaska. Mercury levels were low in these birds compared to other marine species. Overall, we found little evidence that the health and fitness of these animals were affected by exposure to metals. Nevertheless, the following relationships suggested that exposure to metals may affect some aspects of health and fitness: i) mercury was inversely correlated with abdominal fat mass in males and females; ii) cadmium was inversely correlated with abdominal fat mass, total body weight, heart weight and kidney weight in females; and iii) the cell mediated immune response to the mitogen, phytohemagglutinin-P was inversely correlated with cadmium levels in females.

Objectives

Long-term

- To determine whether metals exposure in common eider ducks might be contributing to observed recent, precipitous population declines by damaging their health and fitness.

Short-term

- To determine whether heavy metal levels in these ducks are related to selected measures of their health and fitness, including body condition, immune function and stress hormone response.

Introduction

Northern governments and communities are interested in obtaining information to ensure a sustainable harvest of seaduck species used for traditional/country food. In the Inuvialuit Settlement Region, hunters harvest 2000 to 5000 eiders each year (Fabijan et al. 1997). It has been estimated that approximately 14,000 eiders were harvested each year in Nunavut in the early 1980s (Turner et al. 1996). The Nunavut Wildlife Management Board has ranked seaducks (specifically eiders) among their priority species of concern (Nunavut Wildlife Management Board Research Priorities, Arviat meeting, January 19-23, 1998), presumably because of population declines.

There is growing evidence that seaduck population declines are real. For example, the number of king and Pacific common eiders migrating past Point Barrow, Alaska, during spring migration apparently declined 55% between 1976 and 1994 (Suydam et al. 2000). Population declines have also been noted in eiders that reside near the Greenland coast during the fall (Mosbech and Boertmann 1999). Many migrate to Nunavut to breed each summer. Recent information, from the Rasmussen Basin indicating that king eider populations have declined by around 75% (Gratto-Trevor et al. 1998), supports the above estimates. The reasons for seaduck population declines are unclear. Overhunting, oil spills, increased nest predation, contaminants, and nutritional or physiological factors have been cited as possible factors (Goudie et al. 1994, Environment Canada 1996, Turner et al. 1996). While overhunting is a major concern for king and common eiders, other factors are probably also important in regulating populations of these species (Turner et al. 1996).

Worldwide, there is growing suspicion that population declines or massive die-offs of several species are related indirectly to pollution. The growing list includes several species of birds and marine mammals, for example, striped dolphins from the Mediterranean Sea, bottlenose dolphins from the eastern seaboard of the United States, harbour seals from the North and Baltic Seas, and bald eagles on the Great Lakes (Colborn et al. 1996). The central tenet of this study is that contaminants could be interacting with other physiological stressors in the environment to exert population level effects on eiders in the Canadian Arctic.

There is some evidence that certain metals, including cadmium (Cd) and mercury (Hg), have increased in recent times in northern environments where many seaducks breed and it has been suggested that this phenomenon has resulted from deposition of contaminants transported from temperate zones (Boutron 1995, Lockhart et al. 1995). The problem of contamination of Arctic marine environments has been documented in detail in Canada by the Northern Contaminants Program (Jensen et al. 1997) and by the Arctic Monitoring and Assessment Programme, an international program that provides a global perspective (Arctic Monitoring and Assessment Programme 1998).

Seaduck species overwinter largely outside the Canadian Arctic and return to these areas each spring bearing contaminants accumulated on polluted wintering grounds (Henny et al. 1991). Thus Arctic seaducks, including eiders, may be increasingly exposed to contaminants because of increasing contamination of Arctic breeding areas and southern wintering areas.

Seabirds, including seaducks, have relatively high concentrations of heavy metals. With regard to northern species, this applies to spectacled eiders and Stellar's eiders from Alaska. It is well known that seabirds have relatively high concentrations of certain metals when compared to freshwater or terrestrial species (Di Giulio and Scanlon 1984, Ohlendorf et al. 1991, Henny et al. 1995). Moulting or nesting surf and white-winged scoters and Stellar's eiders from Alaska had high levels of Cd and selenium (Se) and moderate levels of Hg. It is possible that most of the uptake of these contaminants occurs on the wintering grounds (Henny et al. 1991). Recently, Trust et al. (2000) reported that spectacled eiders from Alaska had highly elevated concentrations of certain trace elements, most notably, Se.

There is recent evidence of sublethal effects of Hg, Se and Cd in birds, including seaducks. For example, body weights of wintering seaducks on the Pacific coast were inversely related to Cd (Henny et al. 1991) and Hg (Hoffman et al. 1998). Heart weight was inversely related to Hg in seaducks wintering on the Pacific coast (Hoffman et al. 1998). High concentrations of Se or Hg may alter glutathione metabolism (Hoffman et al. 1998) and could lead to embryotoxicity if a sufficient quantity is transferred to the egg (Henny et al. 1995, Heinz and Hoffman 1998). Although seabirds may be protected from the adverse effects of Cd by the production of the metal-binding protein, metallothionein, some adverse effects of Cd exposure have been documented including nephrotoxicity (Nicholson et al.1983) and altered behaviour in experimental ducklings whose parents were fed Cd before laying eggs (Heinz et al. 1983). There is increasing evidence that contaminants, including metals, can disturb the endocrine status of animals. Hormones play a vital regulatory function in many physiological processes including metabolism, growth, reproduction and thermogenesis. Experimental exposure of fish to Hg (Bleau et al. 1996) and Cd (Hontela et al. 1996) resulted in disturbance to adrenal and thyroid secretions and to carbohydrate metabolism. Fish that are naturally exposed to heavy metals along a pollution gradient in Quebec demonstrated altered levels of adrenal gland secretions; these were linked to a lower bioenergetic efficiency in highly contaminated fish (Sherwood et al. 1997). Hg (Pollard and Hultman 1997), Cd (Boroskova and Dvoroznakova 1997) and Se (Fairbrother and Fowles 1990) can impair the immune system. Impairment of the immune system can increase the susceptibility of animals to disease (Boroskova and Dvoroznakova 1997).

In this study, we are examining selected measures of health and fitness in common eider ducks breeding at

East Bay Migratory Bird Sanctuary on Southampton Island. In particular, we are determining whether relationships exist between tissue concentrations of Hg, Se and Cd and body condition, immune function and corticosterone stress response.

Activities in 1999-2000

A study was carried out from June 15 to July 7, 1999, at the East Bay Migratory Bird Sanctuary on Southampton Island, Nunavut, (64°04′ N 81°40′ W). Twelve male and 12 female common eider ducks were captured. They were immediately blood sampled for hormone analysis, then weighed and measured.

Immune function was assessed in these ducks by examining their responses to phytohemagglutinin-P (PHA-P) and sheep red blood cells (SRBC). The former elicits a simple, rapid cell-mediated immune (CMI) response while the latter promotes a humoral immune response. Two mL of a 20% solution of SRBC in sterile phosphate buffered saline (PBS) were injected intraperitoneally within 2 hours of initial capture. Antibody levels peak about 7 to 9 days after SRBC injection, so the birds were held in a 2.4 × 7.4 × 1.2 m high-flight pen for 8 days prior to blood sampling to assess antibody response. During this period, they were supplied with food (fish) and fresh water ad libitum. On the seventh day after capture, the thickness of both wing webs was measured with a micrometer, then 0.1 mL of PHA-P in PBS (1.0 $mg \cdot mL^{-1}$) was injected intradermally into the right wing and 0.1 mL PBS alone was injected into the left wing web. The swelling response was measured 24 hours later. The response to PHA-P was calculated as the ratio of the post- to pre-injection wing web thickness. The cell mediated immune response to PHA-P was calculated as the ratio of the post- to pre-PHA-P injection right wing web thickness minus the post- to pre-PBS injection left wing web thickness.

A stress response was also determined in these birds by sequentially determining corticosterone hormone levels in blood samples taken sequentially three times over a 30- to 60-minute handling period. This was done twice, once immediately following initial capture and again after being caged for eight days. The three blood samples were drawn at approximately 3 to 5, 10 to 25, and 30 to 60 minutes after capture and handling commenced.

After 8 days, the birds were sacrificed and body condition was evaluated (total body weight, abdominal fat mass, liver mass, kidney mass, spleen mass, heart mass, mass of empty digestive tract). Livers and kidneys were frozen and subsequently analyzed by a combination of cold vapour, and flame and graphite furnace atomic absorption spectrometry for total Hg and Se (livers), and Cd (kidneys). These analyses were done at the National Wildlife Research Centre. Corticosterone levels and total and immunoglobulin G (IgG) antibody titers were determined by radioimmunoassay and hemagglutination titers, respectively, at the University of Saskatchewan.

Results

Metal concentrations

Males had higher levels of Se ($p = 0.0002$) and Cd ($p = 0.05$) in their tissues than females while Hg levels were similar in the two sexes ($p = 0.68$). Geometric mean levels of renal Cd and hepatic Hg and Se in males and females were, respectively, 133 and 81, 32 and 14, and 2.7 and 2.6 $\mu g \cdot g^{-1}$ dry wt (Table 1). Cd was > 300 $\mu g \cdot g^{-1}$ in two of 12 males.

Body condition

Preliminary analyses have shown few significant relationships between metal levels and indices of body condition. In males and females, Hg level was inversely correlated with abdominal fat mass (males: $r = -0.61$, $p < 0.05$; females: $r = -0.68$, $p < 0.01$) (Table 2). In females, body weight at capture ($r = -0.63$, $p < 0.05$), kidney weight ($r = -0.66$, $p < 0.05$), heart weight ($r = -0.70$, $p < 0.01$) and abdominal fat mass ($r = -0.71$, $p < 0.01$) were negatively correlated with Cd level (Table 2). Se levels were positively correlated with liver weight ($r = 0.61$, $p < 0.05$), kidney weight ($r = 0.57$, $p < 0.05$) and heart weight ($r = 0.64$, $p < 0.05$) in females. Of 42 analyses, there were nine significant correlations, a somewhat higher number than would be expected based on chance alone. These data will be further examined using more appropriate multivariate techniques.

Table 1. Geometric mean (95% CI) hepatic mercury and selenium and renal cadmium concentration ($\mu g \cdot g^{-1}$) in male and female common eiders, East Bay Migratory Bird Sanctuary, Southampton Island, June-July, 1999

	Sex	
Metal	**Male (n = 12)**	**Female (n = 12)**
Hg	2.73 (2.06-3.61)	2.58 (2.26-2.94)
Se	32.11 (23.13-44.57)	14.09 (11.22-17.68)
Cd	133.99 (91.33-196.57)	81.15 (55.05-119.64)

Table 2. Pearson correlation coefficients between measures of body condition and log-transformed tissue metal concentrations in male and female common eider ducks

		Metal		
Sex	**Measurement**	**Hg**	**Se**	**Cd**
Male (n = 12)	capture weight	–0.51378	0.34427	–0.20854
	final weight	–0.45211	0.28370	0.00233
	liver weight	–0.09143	–0.01917	0.41114
	kidney weight	–0.06760	0.13296	–0.01072
	spleen weight	–0.50202	0.34776	–0.36660
	heart weight	–0.22345	0.15719	0.48841
	abdominal fat weight	–0.61301*	0.49936	–0.30688
Female (n = 12)	capture weight	–0.12990	0.38788	–0.62674*
	final weight	–0.20750	0.32908	–0.45605
	liver weight	–0.02766	0.60808*	–0.51701
	kidney weight	–0.08259	0.56875*	–0.65899*
	spleen weight	–0.16993	–0.26281	0.56238
	heart weight	0.25719	0.63789*	–0.70472**
	abdominal fat weight	–0.68480**	0.53816	–0.71204**

* $p < 0.05$
** $p < 0.01$

Stress response

Corticosterone levels were determined in blood samples that were taken serially, approximately 3 to 5, 10 to 20, and 30 to 45 minutes after starting to handle each bird. This procedure was done when the birds were initially captured and again after 8 days in the flight pen. Corticosterone levels were affected significantly by interactions between gender and sampling period (at capture versus 8 days following capture) ($p = 0.003$) (Table 3) and between sampling period and serial sample (first, second or third sample during the 30- to 60-minute handling period) ($p = 0.0001$). At capture, corticosterone levels increased from the first to third blood sample (Table 4). However, they remained relatively constant 8 days after capture. At capture, males and females had similar corticosterone levels. However, 8 days after capture, females had significantly lower corticosterone levels than males ($p < 0.05$). Metal levels were not correlated with the corticosterone stress response (difference between concentrations in first and third serial sample), at the initial capture and after 8 days in the flight pen (all p values > 0.05, Table 5).

Table 3. Mean ± 1SE (n) corticosterone levels ($ng{\cdot}g^{-1}$) in plasma samples from male and female common eider ducks*

	Sampling Period	
Sex	**Initial Capture**	**8 Days After Capture**
Male	45.88 ± 3.39 (36)	35.21 ± 2.97 (36)
Female	43.16 ± 2.57 (36)	16.75 ± 1.95 (36)

* Samples were collected immediately capture and after placement in a large flight pen for 8 days.

Table 4. Mean ± 1SE (n) corticosterone levels ($ng{\cdot}g^{-1}$) in plasma samples from common eider ducks*

	Sampling Period	
Serial Sample	**Initial Capture**	**8 Days After Capture**
First	32.45 ± 2.57 (24)	28.62 ± 3.24 (24)
Second	47.26 ± 3.03 (24)	26.69 ± 3.91 (24)
Third	53.85 ± 3.91 (24)	22.62 ± 3.63 (24)

* A series of three samples were collected at approximately 3 to 5, 10 to 25, and 30 to 60 minutes after capturing and handling the birds. This procedure occurred twice, once immediately following the initial capture and again after spending 8 days in a large pen.

Table 5. Pearson correlation coefficients between the corticosterone stress response and log-transformed tissue metal concentrations in common eider ducks immediately following capture and after 8 days in a flight cage*

Metal	**Corticosterone Stress Response Initial Capture**	**Corticosterone Stress Response 8 Days After Capture**
Hg	0.05089	–0.13099
Se	0.36798	–0.06310
Cd	0.11468	0.09875

* The stress response was measured as the difference in corticosterone levels between the third and first blood samples taken at 3 to 5 minutes and 30 to 60 minutes after the initiation of capture and handling.

Immune function

Antibody titers were significantly higher in females than in males ($p = 0.03$, Table 6). Therefore, the relationship between metal concentrations and antibody titers were examined separately according to gender. Total and IgG antibody titers were not significantly correlated with metal concentrations in either sex (all p values > 0.11, Table 7).

The cell-mediated immune response to PHA-P did not differ between the sexes ($p = 0.14$, Table 6). Furthermore, while it was not correlated with metal concentrations in males (all p values > 0.1, Table 7), in females, its relationship to Se concentration was significant ($r = 0.71$, $p < 0.01$), while its relationship to Cd concentration was nearly significant ($r = -0.53$, $p = 0.077$, Table 7).

Table 6. Measures of immune function (median, quartiles) in male (n = 12) and female (n = 12) common eider ducks, East Bay Migratory Bird Sanctuary, June 15-July 7, 1999[a]

Sex	Total Antibody Titer[b]	IgG Titer[c]	Response to PHA-P[d]
Male	7 (4-20)A	10 (2-28)A	0.13 (0.03-0.20)A
Female	32 (8-48)B	24 (6-64)A	0.13 (0.05-0.23)A

[a] Within a column, rows with similar uppercase letters are not statistically different ($p < 0.05$).

[b] Measured as 1/highest dilution factor where hemagglutination was observed.

[c] Measured as 1/highest dilution where hemagglutination was observed in assays using 2-mercaptethanol.

[d] Calculated as: (Post-PHA-P injection right wing web thickness – Pre-PHA-P injection right wing web thickness) – (Post-PBS injection left wing web thickness – Pre-PBS injection left wing web thickness) (mm).

Table 7. Spearman correlation coefficients (r values) between measures of immune function and tissue metal concentrations in common eider ducks

Sex	Immune Function Assay	Tissue Metal Concentration Hg	Se	Cd
Male (n = 12)	Total antibody titer	–0.39	–0.11	–0.42
	IgG	–0.45	0.30	–0.48
	CMI	0.17	–0.10	0.35
Female (n = 12)	Total antibody titer	0.20	–0.14	0.15
	IgG	0.01	0.15	–0.27
	CMI	–0.14	0.71*	–0.53**

* Significant at $p < 0.01$

** $p = 0.077$

Discussion

Tissue metal concentrations

Metal concentrations in this study were similar to those documented in common eider females in 1997 at East Bay (Wayland et al. 1999a). Cd concentrations were higher than those reported in eider ducks at other locations (Karlog et al. 1983, Norheim 1987, Nielsen and Dietz 1989, Hollmen et al. 1998, Trust et al. 2000). However, Se concentrations were lower than those reported in other eider species from western Alaska (Trust et al. 2000). Hg levels were much lower than those reported in other species of marine wildlife (Arctic Monitoring and Assessment Programme 1998, Jensen et al. 1997). Concentrations of hepatic Se and renal Cd were higher in males than in females. Similarly, in Alaska, Cd levels were higher in male Barrow's goldeneye than in females (Franson et al. 1995) while, in Finland, Se levels in male common eiders were higher than in females (Hollmen et al. 1998). Other studies of seaducks have failed to find gender-related differences in metal levels (Karlog et al. 1983, Norheim 1987, Nielsen and Dietz 1989). It is possible that differences between the sexes in seasonal patterns of body and organ weight changes may account for the observed differences in metal concentrations in this study. Females, but not males, undergo dramatic increases in weight in the weeks preceding egg-laying (Gorman and Milne 1971) when this study was done. As tissue weight increases, the concentration of certain metals may become diluted, thus resulting in decreased concentrations but not of total content of metals. Total content of metals in liver and kidneys of these animals will be examined.

Body condition and metals

There were few significant correlations between metal concentrations and various measures of body condition. Nevertheless, Hg and Cd were inversely related to some of our condition measures, a finding that is similar to those reported elsewhere, wherein seaducks with low body weights tended to have relatively high concentrations of Cd or Hg (Henny et al. 1991, Ohlendorf et al. 1991, Hoffman et al. 1998). Fish living on metal-polluted lakes in Quebec were inefficient at converting food energy to somatic tissue, and, as a consequence, were in poor condition (Sherwood et al. 1997). Dietary Cd coupled with a restricted diet enhanced the metabolism of lipids in mallard ducks, thus promoting a drain on stored energy reserves (DiGiulio and Scanlon 1985). However, in this study, it is not clear whether relatively high concentrations of these metals promoted poor condition through mechanisms that affected metabolic efficiency or whether the relatively high metal concentrations were simply the result of lower tissue mass (and thus poor condition).

Stress hormones and metals

The corticosterone response to standardized capture stress was unrelated to tissue metal levels. This is similar to results obtained in 1997 but contrasted with results obtained in 1998 (Wayland et al. 1999b). Corticosterone levels in mallards that were fed Cd have been reported not to differ from those in control mallards (Di Giulio and Scanlon 1985). However, corticosterone levels were higher in mallards that were simultaneously fed Cd and placed on a food-restricted diet, suggesting that Cd could enhance the effects of food restriction on corticosterone secretion.

Fish that are chronically exposed to metals typically are incapable of mounting a strong stress response while those that are acutely exposed secrete higher than normal levels of stress hormones (Hontela 1998). Our data suggest that eider ducks do not respond to metals in the same manner as fish or that the levels of metals to which these ducks were exposed were lower than those required to produce a response.

Immune function and metals

The response to SRBC injection was not related to metal concentrations in either sex. However, the CMI response was inversely correlated with Cd concentrations and positively correlated with Se in females. Cd suppressed T-cell and B-cell production in guinea pigs experimentally infected with ascariasis (Boroskova and Dvoroznakova 1997), and suppressed antibody production in rabbits experimentally inoculated with pseudorabies virus (Koller 1973), indicating that Cd can be immunotoxic. However, there is no published information that chronic low-level exposure to Cd can suppress the immune system. There is a need for further research to determine whether the inverse relationship between the CMI response and Cd concentrations in females eiders is real.

The elevated CMI response in eider females with high tissue Se concentrations was not consistent with the absence of an effect of sodium selenite on CMI response in mallards (Fairbrother and Fowles 1990). However, it was consistent with elevated lymphocyte blastogenesis in the presence of the T-cell mitogen, concanavalin A that occurred in avocets (*Recurvirostra americana*) that were naturally-exposed to relatively high levels of Se, arsenic and boron (Fairbrother et al. 1994). Se at low concentrations is immunostimulatory but at higher concentrations is immunosuppressive (Fairbrother et al. 1994). It is possible that even the higher levels of Se in eider females in this study were in the range that would actually stimulate their immune systems.

In conclusion, we found relatively few relationships between tissue metal concentrations and our various measures of health and fitness. However, the reduced body condition and apparent immunosuppression in eiders with relatively high concentrations of Cd warrant further investigation. We intend to pursue such research in 2000-2001.

Expected Project Completion Date

March 2001

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Monitoring the Health of
ic Peoples and Ecosystems
and the Effectiveness of
International Controls

Part C: Local Contaminants Concerns

Northwest Territories and Nunavut Local Contaminants Concerns

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◆ **Project Team:**

Participating communities; Members of the NWTECC, including representatives from: Dene Nation; Métis Nation-Northwest Territories; Inuit Tapirisat of Canada; Inuvialuit Game Council; Nunavut Tunngavik Inc.; Gwich'in Tribal Council; Sahtu Secretariat; Deh Cho First Nations; Dogrib Treaty 11; Akaitcho Territory Tribal Council; North Slave Métis; South Slave Métis; DIAND; Environment Canada; Department of Fisheries and Oceans; Government of the Northwest Territories (GNWT) Resources, Wildlife and Economic Development; GNWT Health; Aurora Research Institute; and Nunavut Research Institute

Abstract

The Local Contaminants Concerns (LCC) component of the Northern Contaminants Program works with northern communities to resolve local contaminant issues that have a potential to impact on the consumption of traditionally harvested foods. In 1999-2000, the Northwest Territories Environmental Contaminants Committee approved five proposals from communities that had concerns with contaminants at specific sites in their areas. Four of the LCC funded projects were in the Northwest Territories and one was in Nunavut. Information and results for the individual projects are available from the Contaminants Division of the Department of Indian Affairs and Northern Development in Yellowknife.

Objectives

1. To work with northern communities to resolve local contaminant issues that have potential to impact on the consumption of traditionally harvested foods and ultimately human health.
2. To focus on problem definition and assessment with priority given to local contaminant issues identified by communities.

Introduction

The Local Contaminants Concerns (LCC) component of the Northern Contaminants Program (NCP) works with northern communities to resolve local contaminant issues that have a potential to impact on the consumption of traditionally harvested foods. Research under this component provides essential data for informing people about risks and measures to reduce their exposure to risks, as well as for contributing to long-term strategies to control and reduce contaminants. An essential element of this component is the ability to address local concerns. Local contaminant concerns affect the confidence of Northerners with regard to their traditional foods and the health and safety of their environment. In general, the LCC envelope was established to enable small studies to determine if there is a need for further work. If a contaminants related problem is identified through the LCC program, then further work is to be funded through larger programs or regulatory activities, such as the Department of Indian Affairs and Northern Development (DIAND) Waste Program or the NCP.

Activities and Results in 1999-2000

Under the LCC envelope of the NCP there were five projects approved and funded. The NCP provided $100K that was allocated to the projects based on funding determinations made by the Northwest Territories Environmental Contaminants Committee. The LCC projects that were supported in 1999-2000 are briefly described below:

Metal concentration in vegetation of the North Slave Region

This study by the North Slave Métis Alliance investigated the availability of heavy metals in vegetation in the vicinity of six abandoned mines in the North Slave Region. The project proposed to sample lichen and berries but had to scale back to lichens only due to a scarcity of berries at the sites. This project was supported to $27 410 by the NCP.

The final report from this study identified uranium at the Rayrock site as the only parameter that would require further assessment relative to Canadian Council of Ministers of the Environment guidelines to determine if the levels encountered there pose any environmental or human health risk. Currently there is a monitoring program in place at Rayrock that was initiated subsequent to a 1996 site remediation project.

Overall, this study concluded that the abandoned mines posed little heavy metals risk to the environment and human health since the areas affected are small and isolated. It was identified that there could be some transfer to migratory wildlife such as caribou but it was felt to be insignificant. The final report on this study is available through the DIAND Contaminants Division office in Yellowknife.

Risk characterization of arsenic exposure from consumption of berries in the Akaitcho Territory

This study was initiated by the Yellowknives Dene First Nation and included the participation of the Dene Nation. Funding from NCP totalled $26 610 for this project. An earlier study of arsenic in berries in the summer of 1998 recommended further work on the speciation of arsenic and advised caution on Yellowknife area berries and recommended to avoid eating berries from mine sites (Davey 1999). This earlier work gave rise to this risk characterization study.

The draft final report states that arsenic concentrations in soil and berries are higher in the vicinity of the Yellowknife mines. The study concluded that berries can take up some arsenic from soil and suggested that it was safe for an adult to eat up to one cup of berries from local mine sites everyday. Reference is made to earlier dietary studies indicating that most people do not eat enough berries to pose a health concern. The final report should be available soon.

Establishment of baseline arsenic levels for the Yellowknife area to assist in the development of regional remediation guidelines for soils

The Yellowknife Arsenic Soil Remediation Committee submitted the proposal for this study. Approval was given under the LCC envelope and the project was funded for $16 000. In this study, the NWT Branch of the Canadian Public Health Association seeks to determine the origin of the arsenic in archived soil samples and to establish a control site for baseline work away from the influence of local mines. As of May 31, 2000, no results have been reported from this work.

Fish collection and analysis – Peel River at Fort McPherson

The Gwich'in Tribal Council is undertaking this study supported by NCP for $17 501. The concern from Fort McPherson stems from knowledge of the presence of DDT and other possible contamination at the site of an eroding oil exploration disposal pit near the confluence of the Peel and Caribou rivers upstream of the community. The concerns relate to possible occurrence of contaminants in river water and traditionally used fish species. As of May 31, 2000, work is proceeding with no laboratory results reported so far.

Survey and assessment of contaminants at the Nottingham Island Radio Station, Nunavut

This proposal by the Aiviit Hunters and Trappers Association (HTA) of Coral Harbour was approved to a total funding level of $25 000. Integral to the project was an agreement to use the services of Taiga Environmental Laboratory which would enable the utilization of services credits held by the Contaminated Sites Office of DIAND. This arrangement reduced the NCP funding to $12 500 thus keeping the LCC program within budget.

Knowledge of contaminant problems at other abandoned communication and defense installations in the North concerned the Aiviit HTA and led them to initiate and undertake this study. Unfortunately, bad weather during the boat trip to the site for the survey work forced cancellation of the project for the year.

Conclusions

These five studies were community-initiated and addressed contaminant issues that were within the LCC envelope with possible impacts on human health and traditional foods. Final results for the 1999-2000 LCC projects should all be available soon through the NWT Contaminants Division of DIAND in Yellowknife.

The Nunavut Environmental Contaminants Committee was not fully in place in 1999-2000, so the NWT Committee handled Nunavut LCC activity on an interim basis for the year. It is expected that the Nunavut Environmental Contaminants Committee will be operational and dealing with local contaminants concerns for Nunavut during year 2000-2001.

The Local Contaminants Concerns program continues to be an effective way for communities to address specific local concerns regarding contaminants in the environment and traditional foods and the implications this may have for human health.

Expected Project Completion Date

Ongoing

References

Davey, E. 1999. Arsenic levels in berries and soils from the Yellowknives Dene First Nation traditional territory. In: S. Kalhok (ed.), *Synopsis of Research conducted under the 1998/99 Northern Contaminants Program*. Ottawa ON; Indian Affairs and Northern Development:81-85.

Yukon Local Contaminants Concerns Program

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Yukon Contaminants Committee and various other stakeholders for the individual projects

Abstract

The Local Contaminants Concerns (LCC) program completed its second year of operations. Three projects have been selected as representative of program operations. The first was an investigation into the safety of consuming lake trout from Coal Lake. Results to date indicate that trout should be safe for consumption but a full report will await the receipt of additional analyses. The second was a response to concerns raised by a local First Nation regarding the water quality effects of an old mine property in the Macmillan Pass area of the Yukon Territory. A contribution agreement with the First Nation and LCC allowed for a consultant to review the site. The report prompted a meeting between the mining company, the First Nation and the regulatory authority. The mining company has been directed to acquire a licence for the site and to begin clean-up. The third project involved an area contaminated with unexploded ordinance, including an area under land claims selection. The LCC contacted the Department of National Defence for assistance with evaluating the area. This cooperative effort has been conducted with the involvement of the First Nation whose land claim is affected. The Department of National Defence will fully evaluate the area in the summer of 2000; a complete clean-up is planned for 2001.

Objectives

- To address local concerns related to contaminated sites in the Yukon.
- To evaluate individual sites and determine the responsible authority or stakeholder.

Introduction

The Local Contaminants Concerns (LCC) Program in the Yukon, which receives direction from the Yukon Contaminants Committee (YCC), addresses a range of community concerns related to suspected local sources of contamination. In addition to planned projects, a number of studies are initiated annually through "walk-in" contacts from concerned members of the public and other agents. In some years, these walk-in projects represent a significant proportion of the entire LCC program.

In practice, the LCC investigates sites that have been identified through previous planning or the walk-in route. The sites are then investigated in a number of ways. If a site evaluation reveals a level of contamination or a specific contaminant that is in violation of existing legislation, then the LCC program turns the information over to the appropriate regulatory agency for action. If the site is non-regulatory and/or is on lands for which the federal government is responsible, the investigation is turned over to the Department of Indian Affairs and Northern Development (DIAND) Waste Management Program, for action. In instances where there is no contamination found, a report is sent to the initiating individual, organization or agency, and no further action is necessary.

Activities in 1999-2000

A number of projects were conducted during the 1999-2000 fiscal year. Three examples below have been selected as representative of the scope of LCC activities.

Coal Lake

Description

A walk-in contact by a member of the public and a local outfitter raised a concern regarding the safety of consuming Lake Trout from Coal Lake. The concern related to the food fish consumption advisory on Lake Laberge, which is nearby. As Coal Lake is part of the international Wolf Creek Watershed Project administered under the Water Resources Division of DIAND, previous work on the site had provided organochlorine analysis of sediment cores and benthic species. Lake Trout were collected in July 1999 to supplement this data and directly address the consumption issue. Tissue samples were removed from the fish and sent to the Department of Fisheries and Oceans laboratories in Winnipeg for organochlorines and metals analysis.

Results

The complete results from the laboratory have yet to be received, but a review of limited previous sampling done at Coal Lake in 1995 indicated that there was no reason for health concerns associated with consuming fish from that lake. When the full results are received, a compilation of results of the 1999 and 1995 sampling will be made in a report format. This report will be sent to the individual and the outfitter who initiated the investigation.

Macmillan Pass (Tom Mine Property)

Description

The Ross River Dena Council (RRDC), who represent the First Nation members in the Ross River region of Yukon, contacted the LCC with concerns about the effects on water quality of drainage from an old mine site within their traditional territories. The records of past activity at the site were located in DIAND files and reviewed. The site is the location of an exploratory adit opened by the Hudson Bay Mining and Smelting Company in 1982. The site is located in a remote area near the border of the Yukon and Northwest Territories. The area is known for naturally occurring acid rock drainage (ARD) and has been the subject of a number of published research papers (Jack and Osler 1983, Kwong and Whitley 1992).

Results

A contribution agreement was arranged between the LCC and the RRDC, who in turn hired a consultant to conduct a site investigation. A review of the existing literature and a new sampling program, undertaken with direction from the First Nation, produced analytical results for water and sediment indicating that the concentration of the ARD drainage from the site has become slightly more acute since the last sampling series in 1991. While the local watershed is already subject to significant natural ARD effects, the drainage from the open adit is above regulatory limits. A report summarizing these findings (Burns and de Graff, 1999) was produced and delivered to RRDC by the consultant.

The report and background details were sent to the Water Resources Division of DIAND, who contacted Hudson Bay Mining about the site. A meeting was arranged in which all stakeholders could discuss the study. The meeting resulted in Water Resources directing Hudson Bay Mining to seek a water licence for the site and to secure the adit. The licence will require effluent treatment at the site to achieve compliance with existing regulatory statutes.

Lake Laberge unexploded ordinance

Description

In the 1950s, the Department of National Defence (DND) employed several areas around and in Lake Laberge as a gunnery range. Local people living along the lakeshore contacted the LCC regarding unfired ammunition found along the shore. Additional information indicated unexploded munitions on Richtofen Island (in Lake

Laberge); significant shrapnel also exists in the area adjacent to Jackfish Bay (also on the lake). This area is under land claims selection by the Ta'an Kwach'an First Nation. A local individual also had underwater video footage of what appeared to be bombs on the bottom of the lake off of Richtofen Island.

Results

The LCC contacted the Department of National Defence (DND) in Yellowknife for expert advice on how to conduct an evaluation of the area. A DND staff member travelled to Whitehorse after reviewing the archival references, video, still photos and shrapnel provided by the LCC, to conduct a field reconnaissance of the area with LCC staff and representatives of the Ta'an Kwach'an. The DND staff member indicated that the underwater objects were practice bombs; the shrapnel had come from 500 or 1000 pound bombs and one piece of recovered material was an unexploded anti-aircraft shell. Subsequent to this, the LCC hired an underwater camera for further reconnaissance of the area adjacent to Richtofen Island and more bombs were located.

DND staff informed their headquarters and a large reconnaissance is planned for the summer of 2000, with a full clean-up planned for 2001. The LCC will continue to assist with logistics and to liaise with the First Nation, media and local community.

Discussion and Conclusions

The LCC program is continuing to provide a cost effective response to local concerns regarding environmental contaminants that have no other ready avenue of approach, while meeting the directives of the YCC. The YCC has allowed the LCC a significant degree of flexibility in responding to concerns as they are raised. In this second year of the program, the LCC has continued to expand the range of issues it addresses.

The LCC has continued to receive good support from the general public, First Nations, and other government agencies. To date, all of the projects undertaken have produced an identifiable conclusion, usually within an acceptable time frame. The challenges for the LCC are meeting the increasing demand and providing a comprehensive response in a timely manner as more stakeholders become aware of the LCC program.

References

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Education and Communications

Elders/Scientists Retreat: Strengthening the Ties in Denendeh (IV)

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Abstract

Communication was the theme for the fourth annual Elders/Scientists Retreat held near Dettah, Denendeh, March 28-31, 2000. Dene Elders and scientists from the Northwest Territories and southern Canada defined communication, explored ways to work together and made recommendations for incorporating traditional knowledge in Northern research. The retreat was attended by 13 Dene Elders, nine scientists, interpreters, a camera operator, a facilitator and the Dene Nation Lands and Environment staff. This diverse group was brought closer together through discussions, meals and evening activities. Topics that were addressed ranged from what traditional knowledge is and how it is reported to how it should be incorporated into Western science and how Elders wish to be included in research. Scientists were open to such discussions and were eager to learn more about how traditional knowledge is collected and how to use it in their research. Many recommendations were made during the retreat, including the need for more education, specifically educating scientists about the Dene ways and traditional knowledge, and educating the youth in Dene ways as well as in Western scientific ways. Other recommendations focussed on the need for improved communication between Elders and scientists, the inclusion of Elders and youth in research, the need for more community meetings concerning research being conducted, and making information from research available to the communities.

Objectives

1. To continue to build on the progress made at the first three Elders/Scientists retreats.
2. To promote a better community dialogue between Denendeh communities and scientific and non-scientific researchers and government and non-government agencies.

Introduction

Traditional knowledge is gaining respect in the scientific community, but there are still many who do not understand what is meant by the term 'traditional knowledge' or how to collect and use this knowledge. The key to understanding traditional knowledge and working together is communication. The Elders/Scientists retreats have promoted community dialogue among Denendeh communities and scientific and non-scientific researchers.

Communication was the theme for the fourth Elders/ Scientists Retreat, which was intended to build upon the trust and respect developed through the previous three retreats. At the fourth retreat, Elders and scientists defined communication, explored ways to work together and made recommendations for incorporating traditional knowledge in Northern research.

Activities in 1999-2000

The retreat was held from March 28-31, 2000 at a facility located near Dettah, Denendeh, just outside of Yellowknife. The facility provided a quiet, relaxed setting where participants had the opportunity to become acquainted and share their experiences. Questions, concerns, information and ideas were openly discussed.

The retreat was attended by 13 Dene Elders, nine scientists, three interpreters, one camera operator, a facilitator, and the Dene Nation Lands and Environment staff (Appendix). Having the staff, interpreters and camera operator stay at the facility with the participants created a feeling of trust among the group and added to the overall success of the retreat. Large and small group discussions, as well as walks, sharing of meals and an evening of drum dancing and Dene hand games, all contributed to bringing the group closer together.

The retreat was loosely structured around a series of questions: "What do you expect from the retreat?"; "How do Elders want to be included and acknowledged in research and how should information be returned to their communities?"; "How are contaminants perceived?"; "What are your past experiences with scientists?"; "What is traditional knowledge?"; "How can we incorporate traditional knowledge and Western science?"; "How does traditional knowledge get reported and acknowledged?"; and "How would Elders wish to change the traditional knowledge reporting and maintain ownership of it?" Each question led to broad and extended discussions, the sharing of experiences, and more often than not, several further questions.

Results

Many questions were raised and addressed during the retreat. These questions were posed to both Elders and scientists as a means of understanding the different outlooks that these two groups hold, as well as providing scientists and Elders with an opportunity to communicate with one another.

Expectations of retreat

All of the participants expressed a willingness to listen, to share and to learn from other participants, and expressed their hopes of enjoying their time at the retreat. In addition, scientists expressed the desire to communicate with the Elders – through conversation rather than through presentations – and to "absorb" as much traditional knowledge as possible. They expressed a desire to incorporate that traditional knowledge into scientific research, but acknowledged that the reality for the scientist is that it is difficult endeavour (for logistical and financial reasons, among others), and therefore some guidance and understanding on the subject would be beneficial.

Elder acknowledgment, inclusion in research, and the return of information

The discussion surrounding the question of how Elders want to be acknowledged and included in research and how information is returned to communities revealed that there are still some difficulties that exist. Although the traditional knowledge of Aboriginal people, as well as the 'local knowledge' of all people who live off the land, is valued and respected, from the scientists' point of view there exist problems of time and finance constraints when it comes to accessing and using this knowledge in research. Scientists use numbers to examine details while Elders have knowledge of 'the big picture' and share their information without the use of numbers. There are opportunities and benefits to making use of both sets of knowledge, for example, Elders may know where to go to look for soil contamination and scientists have the tools to document it quantitatively to present to communities, regions and relevant government authorities. Elders emphasized that Aboriginal people need training on pollution and contaminants, and that scientists need to be educated on the importance of traditional knowledge as well as other traditional activities in order to improve communication between scientists and communities. Also, a suggestion was made for scientists to work with interpreters to develop terminology that is understandable.

Communication between Elders and scientists

Both Elders and scientists agreed that there is a need to improve communication between scientists and communities, and the keys to this lie in better understanding of each other's basis for knowledge (e.g. traditional knowledge or scientific knowledge), mutual respect, and open two-way lines of communication.

Elders have had varied experiences with scientists in the past. Elders emphasized the need the need for scientists to protect animals and noted a lack of respect from mining exploration teams, for example, by going on peoples' trap-lines. Other scientists have shown a lack of respect by not informing a community why they are there and what they are doing. When scientists conduct research in a community, it is of utmost importance (and respect) to report findings back the community – in plain language. Scientists emphasized that most researchers have good intentions, but they may find it difficult to communicate with Elders and the information that they wish to communicate is lost.

Traditional knowledge

Both Elders and scientists shared their ideas of what is 'Dene Traditional Knowledge'. They said that traditional knowledge is the rich heritage of living and surviving on the land, and includes all aspects of respect and knowledge of the land, the air, and other people. Spirituality, land skills, plants, animals, weather and medicine are all aspects of traditional knowledge. It is also the history of the Dene people and the land, told in their own language and with their own stories. These stories came from their ancestors and have been passed on through generations from grandparents to grandchildren. Traditional knowledge is learned best out on the land, hunting and canoeing, learning to live off the land. Skills to survive off the land, such as using tools from branches to make fish nets, as well as discipline and respect, were passed on from Elders to youth so that they would know how to live. Elders know so much about everything, it is often difficult to ask questions and tap into all of their knowledge; they will only tell so much at one time. This means that to understand the traditional knowledge of the Elders, we must have a good and respectful working relationship.

In the last 50 years, the whole planet has moved so quickly, and so much has changed so that many people are puzzled. For the Dene, traditional knowledge is the stabilizing influence and the basis for the future. In the past, money had no value. People helped one another. Youth used to be healthy, which came from the connection of mother to child through breastfeeding. Food used to be mostly traditional, as there was very little market food. Hunting and fishing were a large part of their life, and they were very content to live on the land. Now, it is difficult to live in two cultures. People are out on the land less and less, even though they know that there is a difference between the value of traditional food and food purchased at the grocery store. The weather is changing. It is believed that development and government help is affecting traditional lifestyles and there is a worry that the traditional lifestyle will be lost. Now, with life being 'easier', maybe children should learn about the land and about their language through books.

Elders wish to speak with scientists, and feel comfortable speaking with them as a way of educating them on the importance of traditional knowledge and culture. They suggested that it may be worthwhile to spend time with scientists in the bush.

Integrating traditional knowledge and Western science

The participants all agreed that integrating both traditional knowledge and Western science would benefit all involved. Both Elders and scientists see that by combining the two it will make them stronger. Scientists recognized that they must find a way to use traditional knowledge or the "big picture" first. However, if scientists want to study animals, they need to ask the Elders first. For example, if moose are needed for a study, ask the Elders and community members and they will decide how many and where they should go; Dene laws should be respected. It is believed that scientists could learn much more by living off the land with Elders, they would learn about weather, hunting and trapping. As well, opportunities to take the youth out onto the land are needed, this would incorporate outdoor/traditional life with the Western style of education.

Reporting and acknowledging traditional knowledge

Currently, traditional knowledge is reported and acknowledged through funded traditional knowledge report projects from designated groups or communities; it is referenced within pages of reports as "quotes," or "anecdotal evidence". Most of these reports have been sent to the "South" and nothing has been reported back to the community; it is only through word of mouth that communities hear anything of these reports. Elders wish to change the reporting of traditional knowledge and wish to maintain ownership of it. This could be done by ensuring reports stay in the communities, band offices, and schools, and they believe that the copyright should be to the report authors.

Discussion and Conclusions

The Fourth Dene Elders/Scientists Retreat was filled with sharing of experiences and discussions on traditional knowledge, scientific research, and how these two kinds of knowledge and their holders interact. Discussion on a series of questions worked towards gaining a better understanding of the different kinds of knowledge and an improved relationship between Elders and scientists.

From these discussions, several recommendations were made. First, all who participated agreed that it is important for the youth to have the opportunity to attend retreats of this nature, because they have much to learn, particularly from the Elders, and are the individuals who will work together with and bridge the gap between communities and scientists in the future. Unfortunately, there were not sufficient funds to allow for youth to fully participate in the retreat. Some youth from the community of Dettah were able to attend the evenings of drum dancing and hand games. Every effort to include the youth will be made for future retreats and workshops. Second, education is vital, particularly educating scientists about the Dene ways and traditional knowledge, educating the youth in the Dene ways, and educating the youth in Western science. Third, communication should be a main focus for improvement. More specifically, Elders want to be included and informed about research. Improved two-way communication, with each party recognized as an equal, will help to incorporate Dene knowledge and Western science into research projects. Fourth, more community information meetings where Elders can voice concerns are needed before studies begin as well as upon their completion, especially since communication between Elders and scientists has been lacking once scientists have returned to the "South". Finally, information from workshops needs to be made available to the communities as well.

Acknowledgments

The Dene Nation wishes to thank the Elders, scientists, facilitator, youth, interpreters and camera operator for their participation in the retreat. In addition we would like to thank the retreat planning committee at the Dene Nation for their hard work and active participation in this retreat. Special thanks to the following people for contributing to this project: Dene National Chief Bill Erasmus, Shelly Anderson and Joe Jack, Dene Nation; Dettah Drummers; and the staff at the Northern Addictions Centre. Sincere thanks and appreciation to the following organizations that, with their support and funding, made this retreat possible: the Northern Contaminants Program, the Northern Addictions Centre and Native Communication Services.

Project Completion Date

March 31, 2000

Appendix

Participants in the *Strengthening the Ties in Denendeh IV* Elders/Scientists Retreat, March 28-31, 2000

Elders

Gwich'in

Elizabeth Colin, Fort McPherson
Mary Kendi, Aklavik

Sahtu

Rosie Grandjambe, Fort Good Hope
Victor Meanacho, Tulita

Deh Cho

Albert Bonnetrouge, Fort Providence
Caroline Bonnetrouge, Fort Providence

Dogrib Treaty 11

Alexie Arrowmaker, Snare Lakes
Elizabeth Arrowmaker, Snare Lakes
Elizabeth MacKenzie, Rae-Edzo
Joe Migwi, Rae-Edzo

Akaitcho Territory

Alfred Baillargeon, Dettah
Pierre Marlow, Lutsel k'e

Scientists

Birgit Braune, Canadian Wildlife Services, Environment Canada, Hull, Quebec
Sarah Kalhok, NCP Secretariat, Hull, Quebec
Marianne Kardash, Contaminated Sites Office, DIAND, Yellowknife
Janine Murray, NCP Secretariat, DIAND, Hull, Quebec
Erica Myles, GNWT Health and Social Services, Environmental Protection, Yellowknife
Carol Reynolds, Finance Manager, NCP Secretariat, DIAND, Hull, Quebec
Karen Tofflemire, Inuvik Regional Health and Social Services Board, Inuvik

Guest Participants

Judy Farrow, Métis Nation-Northwest Territories
Miriam Tyson, Métis Nation

Translators

James Marlowe, Chippweyan
Phillip Rabesca, Dogrib
Laura Tutcho, North Slavey

Staff

Allison Armstrong, Dene Nation
Greg Bailey Hopf, Dene Nation
Mariah Itsi, Dene Nation
Peter Squirrel, Camera operator, Native Communication Services
Norman Yakeleya, Facilitator, Northern Addiction Services

Yukon First Nations Dietary Conference

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◆ **Project Team:**

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Abstract

A conference on Dietary Benefits and Risks " Traditional Food: Is it Safe?" was held from March 1-2, 2000, in Whitehorse, Yukon. It was sponsored by the Council of Yukon First Nations (CYFN), in conjunction with the Yukon Aboriginal Women's Council and the Centre for Indigenous Peoples' Nutrition and Environment at McGill University. This conference was attended by approximately 70 to 80 people from all Yukon First Nations' communities. The target group for the conference consisted of the frontline workers in communities (Community Health Representatives, Renewable Resource Council Members and Band Resource Officers), as well as community members. The conference is part of the overall communication strategy set out by the CYFN that began in 1995 in attempts to educate community members at the grassroots level about contaminants. It was at this time that CYFN-Northern Contaminants Program began to work closely with the Community Health Representatives. Two key questions that were raised repeatedly by Yukon First Nations concerned whether our traditional foods are safe to eat and the reason that chronic disease are on the rise.

Objectives

1. To communicate nutritional, contaminants and socioeconomic results reported in *Yukon First Nation's Assessment of Dietary Benefit/Risk* (Centre for Indigenous Peoples' Nutrition and Environment 1998), as well as to exchange information about various risk factors of chronic diseases identified in the report and of concern in Yukon First Nation communities.
2. To provide a forum for continuing the exchange of health-related information as it relates to the use of traditional/country foods.
3. To improve the understanding of the relationship between traditional/country and market food use and human health.
4. To promote informed decision making by individuals and communities in their food use.
5. To continue to build the knowledge and capacity of individuals and communities in the Yukon to integrate and apply nutritional and contaminants information into healthy decision making.

Introduction

The risks associated with the presence of contaminants in Yukon traditional/country foods are low, especially with respect to the heavy metals and organochlorines studied. People who consume a lot of trout and salmon have usual intakes that exceed the tolerable daily intakes of chlordane and toxaphene. Although the risks associated with these levels of exposure are low, it is important that these two fish species warrant continued monitoring, not because of high contaminants but, rather due to the fact that these traditional food species tend to be consumed in large quantities.

In 1995, the Council of Yukon First Nations (CYFN) requested that a dietary study be undertaken by the Centre for Indigenous Peoples' Nutrition and Environment (CINE) to get a better understanding of the kinds of traditional and store-bought foods people were eating, and what the contaminants and nutrients were in those foods. The study was designed and implemented by CINE in collaboration with community representatives and CYFN, and was guided by community research agreements.

Communication of the results and building on them in partnership with communities is an important aspect of the study. Several workshops were held in different communities last year as a means of exchanging information. This year a dietary conference was held to build on the results of CINE's dietary study and to educate the frontline workers, as they are the resource workers in the communities best equipped to get the knowledge to their people.

The conference is part of an overall communication strategy set out by the CYFN to further educate community resource workers at the grassroots level about contaminants. Two issues that Yukon First Nations were concerned about were to be addressed at the conference: the safe consumption levels of traditional foods and possible reasons for the rise in chronic diseases and their relation to contaminants.

Activities in 1999-2000

The conference on Dietary Benefits and Risks "Traditional Food: Is it Safe?" was held March 1-2, 2000, in Whitehorse, Yukon. This conference was well attended by approximately 80 people from all Yukon First Nations communities. The target group for the conference consisted of the frontline workers in the communities, including Community Health Representatives (CHRs), Renewable Resource Councils (RRCs) and Band Resource Officers, as well as other community members.

CHRs and RRCs have been identified as community members who would benefit by having a frontline training package on contaminants developed so that they are better equipped to inform their communities about contaminants and the Northern Contaminants Program.

Women were identified in the CINE Report as an important target group to whom to emphasize the benefits of traditional/country foods, including the increase in physical activity while acquiring and preparing these foods, and their high nutritional value, for example as excellent sources of calcium and vitamin A.

Youth were identified in the CINE Report as being the group of lowest consumers of traditional food, which may increase their risks for chronic diseases, such as diabetes, heart disease and cancer, in the future.

Results and Discussion

A dietary study by CYFN-Northern Contaminants Program and CINE was commenced in 1995 and was completed in 1998. Ten communities participated in the study. The findings of the research confirmed that the diet of Yukon First Nations is rich in traditional food. Traditional food consumed brings about better diet quality and increased physical activity. It was noted that the consumption of traditional foods has decreased among First Nations living in urban areas and among 20- to 40-year-olds in comparison to persons from older age groups.

Many individuals at the March 2000 conference on dietary benefits and risks reported eating less traditional food now than they did five years ago. The reasons for the decrease are lack of accessibility, lack of time, lack of resources to go hunting and animal scarcity. From this we concluded that the lack of traditional food consumption was a result of people living away from their rural communities. The participants also came to the conclusion that the problem of being overweight is common and increasing among the Yukon First Nations women, the elderly and the groups for whom chronic disease (heart disease, diabetes) seem to be on the rise. It is recommended that immediate attention be paid to educate people about improving diet quality and increasing physical activity. An inadequate intake of calcium and vitamin A were noted for all age groups and additional dietary sources (i.e. increased consumption of moose, caribou, fish livers, kidneys and broth from boiled bones) are essential.

Conclusions

The risks associated with the presence of contaminants in Yukon traditional foods are low, especially with respect to the heavy metals and organochlorines studied. People who consume a lot of trout and salmon have usual intakes that exceed the tolerable daily intakes of chlordane and toxaphene. Although the risks associated with these levels of exposure are low, it is important that these two fish species warrant continued monitoring, not because of high contaminant but, rather, due to the fact that theses traditional food species tend to be consumed in large quantities.

In all communities, the benefits of traditional food outweigh the risks. Nevertheless, it is important to stress that the quality of traditional food species needs to be constantly monitored. This is particularly true for species that are consumed most by Yukon First Nations such as salmon, trout and moose.

Expected Project Completion Date

March 31, 2000

References

Centre for Indigenous Peoples' Nutrition and Environment. May 1998. *Yukon First Nation's assessment of dietary benefit/risk*. Ste-Anne-de-Bellevue, Quebec: McGill University.

Yukon First Nations Traditional Knowledge Research Guidelines (Phase II)

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Abstract

The purpose of developing traditional knowledge research guidelines is to ensure that traditional knowledge is given the same weight as scientific knowledge where appropriate, and to help facilitate a positive working relationship that enhances or builds upon respect, trust and cooperation.

Objectives

1. To develop research guidelines on the collection, storage, access, use and ownership of traditional knowledge, for use by Yukon First Nations, scientists and others, and to assist in incorporating traditional knowledge into research projects on health and contaminants.
2. To facilitate cross-cultural understanding.
3. To ensure that proper protocols, which are for the benefit of communities, are followed when scientists access traditional knowledge.

Introduction

There is growing recognition that traditional knowledge contributes to program planning and has the ability to validate western scientific approaches to research, planning and policy development. The use of traditional knowledge along with Western science in monitoring the health of people and the ecosystem has become increasingly important. Traditional knowledge provides information on the uses of plants, natural medicines, and on wildlife and plant harvesting methods.

A traditional knowledge research guideline can enhance the relationship between Western scientists, various stakeholders and Yukon First Nations, and ensure that proper protocols are followed when scientists conduct research involving Aboriginal people, their knowledge and their lands.

The *Traditional Knowledge Research Guidelines* is a guide for researchers or scientists when seeking to access Yukon First Nation traditional knowledge. These guidelines set out methods for the collection, storage, access, use and ownership of Yukon First Nation traditional knowledge. The guidelines are intended to ensure positive working relationships between the Council of Yukon First Nations and researchers, and reflect the unique ways of respecting, protecting and connecting with traditional knowledge.

Activities in 1999-2000

The Council of Yukon First Nations organized a series of consultations from 1997 to 1999 with community members, government agencies and researchers to discuss ways of developing protocols for access to and responsible use of traditional knowledge.

Initial draft guidelines were developed in July 1999. These guidelines were revised a number of times by the CYFN Northern Contaminants Program (NCP) Coordinator (Cindy Dickson), Traditional Knowledge Coordinator (Robert Lee Jackson) and Traditional Knowledge ad hoc Committee. The guidelines were revised to meet the intent of the NCP. Yukon Elders shared the knowledge of the land, water, air, animals, birds, fish, and their comments on contaminants in the environment and the food chain.

A meeting was held in Whitehorse, Yukon, at the Council of Yukon First Nations office in November 1999. The objective of the meeting was to review the draft guidelines that were developed. Forty-four people attended the two-day meeting. They represented Yukon First Nations, Umbrella Final Agreement Boards and Committees, Yukon College, Yukon Archives, and Yukon and Federal government departments. All who attended were pleased that the "draft guidelines" were a reality and that they would be able to review and revise them.

The consultations with the First Nations resulted in three additional topics to be included in the draft guidelines: a sample community research agreement, a sample checklist, and a sample informed choice form.

Results and Discussions

The traditional knowledge guidelines are intended to provide overall instruction and direction for various key concerns regarding traditional knowledge (e.g. collection, storage, access, use and ownership). The guidelines tend to be generalized statements, which may appear overwhelming to communities, but which also provide a basic checklist of steps to follow so that projects can address concerns individually and ensure that each guideline is being followed.

Under the auspices and support of the Council of Yukon First Nations, the Northern Contaminants Program organized a series of roundtable consultations in 1998-1999. Community members, government agencies and researchers attended to discuss the ways and means of developing protocols for ethical access to, and responsible use of, traditional knowledge. This report reflects the outcome of the work sessions and committee meetings held within that time frame.

The traditional knowledge guidelines were presented to the Elders Council on March 13 and 17, 2000, and were tabled with the Council of Yukon First Nations Leadership on March 27, 2000. The CYFN Leadership adopted the *Traditional Knowledge Research Guidelines*; they will be made available to NCP researchers, managers, Aboriginal partners, and those who request a copy of the booklet.

Expected Project Completion Date

March 31, 2000

Frontline Training for Northern Community Professionals

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Abstract

Contamination of the Arctic ecosystem is of concern to all northern Aboriginal peoples. Two-way communication between communities and scientists about contaminants issues is needed, however, there are barriers to this effective communication, including language and culture. Community professionals such as Community Health Representatives or Renewable Resource Officers can be key people in bridging this gap, but they must first be provided with clear, consistent information. The Frontline Training Course for Community Professionals took place in Rankin Inlet, Nunavut, at the Siniktarvik Hotel, January 25-27, 2000. Twenty-one participants and one interpreter attended the course. Northern community professionals from Kivalliq communities of Arviat, Coral Harbour, Chesterfield Inlet, Baker Lake, Whale Cove and Rankin Inlet were represented. The Regional Contaminants Coordinator for the Baffin region also attended. There was excellent participation throughout the three days in class discussions and group activities.

Objectives

Long-term

- To build capacity in all northern communities with respect to basic knowledge of contaminants issues, knowledge of resources in relation to contaminants, and evaluation and negotiation of research proposals.

Short-term

- To hold a three-day course on contaminants and research issues. Target participants will be frontline community professionals such as Community Health Representatives, Renewable Resource Officers, Hunters and Trappers Organization managers, and staff of local Aboriginal organizations.

Introduction

Contamination of the Arctic ecosystem is an issue of concern to all northern Aboriginal peoples since subsistence hunting and fishing is an integral part of the culture. Unexpectedly high levels of contaminants have been detected in Arctic ecosystems, and many of these contaminants can bioconcentrate in animals at the top of the food chain. The health of Arctic fish and wildlife are of primary concern to northern communities. This is a concern that is also shared by scientists in the Northern Contaminants Program (NCP).

Northerners and scientists both have information relevant to contaminant issues. Northerners should have the opportunity to consider both scientific and traditional knowledge before making decisions about traditional food consumption, and traditional knowledge and local concerns should help steer the direction of scientific research. Two-way communication about contaminants issues is needed.

A three-day frontline training course on contaminants issues has been offered to northern community professionals of all regions of Nunavut. The Métis Nation of the Northwest Territories and the Department of Indian Affairs and Northern Development (DIAND) developed this course for several reasons. One of the key objectives of the NCP is to participate in two-way communication with Northerners about contaminants. However, there are barriers to effective communication between communities and scientists, including language and culture. Community professional such as Community Health Representatives or Renewable Resource Officers can be key people in bridging this gap. They must first be provided with clear, consistent information.

The NCP Frontline Training Course is designed to build capacity at the community level. Key people in the community such as Community Health Representatives and Wildlife Officers must be familiar with contaminants issues. Course participants acquire the knowledge to answer or to access information with respect to questions and concerns about contaminants from local residents. These residents are more likely to trust and understand respected community members than visiting scientists. In addition, course participants learn about the rights of Aboriginal people with respect to research and discuss opportunities for local and regional participation in research activities. The three main topics addressed in the course are: i) contaminant information; ii) scientific research; and iii) the proposal process.

The Frontline Training Course was pilot tested in 1997-1998 in Iqaluit, Nunavut and Aklavik, Northwest Territories. If was offered in 1998-1999 for participants from the Kitikmeot region and most recently, in January 2000, for participants from the Kivalliq region. After each course, revisions were made based on recommendations from the participants and instructors, and course material was modified to ensure that it is up-to-date and regionally relevant.

Activities in 1999-2000

In the fall of 1999, consultation was undertaken in the Kivalliq region to seek the cooperation and support of the Nunavut Department of Health and Social Services, the Kivalliq Regional Health and Social Services Board, and the Department of Sustainable Development, Wildlife and Environmental Protection, Arviat.

The Frontline Training Course for Community Professionals took place in Rankin Inlet, Nunavut, at the Siniktarvik Hotel, January 25-27, 2000. Lena Selamio, the Regional Contaminants Coordinator from Tuktoyaktuk, was the assistant instructor. Her experience as Regional Contaminant Coordinator in the Inuvialuit region and as a member of a northern community, as well as her previous experience in social work, were a considerable asset. Miriam Tyson, Métis Nation-Northwest Territories Contaminant Education Coordinator, who delivered the course for Kitkimeot participants in November 1998, was the chief instructor and did the major revisions to course material for the Kivalliq region.

Prior to the course, materials were reviewed to incorporate recommendations from the workshop held in November 1998. Some sections were updated to include recent information and studies relevant to the target region. The nutritionist for Nunavut, Jill Christensen, reviewed the nutrition activities and provided nutrition fact sheets to be distributed to participants. Erica Myles of the Government of the Northwest Territories Health Department provided suggestions for the health hazard assessment activities. Glen Stephens and David Kennedy of DIAND Contaminants Division, Yellowknife, assisted in the preparations by providing texts for the course and advice as requested.

Based on the experience of past Frontline Training courses and recommendations of past participants, some modifications were made for the January 2000 workshop. These included the following:

- The course took place over three days instead of two, and the duration of each day's session was shortened. The amount and intensity of material presented, along with the fact that many participants were not used to sitting inside for a whole day, made the shortened days appropriate.
- Overheads used to present NCP information were modified to include more pictures and fewer words.

- Activities and overheads involving the NCP Project Summary Booklets were updated to use 1999-2000 projects.
- Activities involving research studies were modified to include studies that were relevant for the Kivalliq region. Studies that were relevant across the North were kept.
- Overheads and activities for the health hazard assessment section were modified, again to include more pictures and fewer words, and to allow participants to see, step by step, how the recommended maximum weekly intake (RMWI) is calculated.
- New activities and materials were added in the food chain section and the scientific research and NCP proposals sections. These activities were incorporated from the Regional Contaminant Coordinators Workshop, the Northwest Territories Science Curriculum and the Yukon Contaminants Curriculum documents.

Logistics for the course were arranged from the Métis Nation-Northwest Territories office by Nancy Peel, who is a Métis from the territory and a student at Aurora College in Yellowknife.

Results

Twenty-one participants and one interpreter (one of the participants acted as a second interpreter for part of the course) attended the Frontline Training course. Northern community professionals from Kivalliq communities of Arviat, Coral Harbour, Chesterfield Inlet, Baker Lake, Whale Cove and Rankin Inlet were represented. The Regional Contaminant Coordinator for the Baffin region, Stephanie Yung-Hing, also attended. Occasional observers dropped in during the course presentation.

Discussion and Conclusions

There was excellent participation throughout the three days in class discussions and group activities. Perceptive questions and comments by many participants showed that some topics were well understood. All participants also completed an evaluation form. On the whole, participants enjoyed the course and found that it progressed at a reasonable pace, with good balance between activities and presentation.

The following recommendations should be considered for any future Frontline Training course:

- Frontline training should be offered to the Dene and Métis of the Northwest Territories, the only group in Nunavut and the Northwest Territories that has not received this training.
- Technical terms continue to be a difficulty for many participants. Extra effort should be made to make sure definitions are clear whenever technical terms are used. Important definitions should be posted on the wall in the room where training is taking place.
- The manual and materials should be revised to incorporate suggestions by the participants and instructors, and prepared so that they may be transferred to other organizations for delivery.

Project Completion Date

A Frontline Training Course is planned for delivery community professionals in Dene and Métis communities of the Northwest Territories in 2000-2001.

Training and Support for Regional Contaminant Coordinators

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Abstract

Regional Contaminant Coordinators (RCCs) work with northern communities to facilitate two-way communication and capacity building for dealing with contaminant issues. A training workshop was held September 27-28, 1999, in White Rock, BC, to provide the RCCs with an overview of the scope and findings of the Northern Contaminants Program so that they may, in turn, provide consistent messages to communities with regard to research results and other Program related activities. Participants included RCCs from the Gwich'in, Inuvialuit and Labrador regions. The workshop covered many aspects of an RCC's responsibilities, including evaluating communication priorities, developing educational materials, organizing workshops, time and budget management strategies, and proposal writing and evaluation. It also provided an opportunity for RCCs to get to know one another and to share ideas, experiences and knowledge. Participants expressed some of the difficulties they face in their positions and made recommendations on how to improve communications between themselves, communities, Aboriginal organizations and the Northern Contaminants Program. They also provided recommendations for future training workshops, including a recommendation to extend the course over additional days in order to adequately cover the course material.

Objectives

1. To provide Regional Contaminant Coordinators (RCCs) with an overview of the scope and findings of the Northern Contaminants Program (NCP) so that they may, in turn, provide consistent messages with regard to research results and other Program related activities.
2. To provide RCCs with training and support so that they may carry out their work in their respective regions.

Introduction

The role of RCCs is essential for the NCP to accomplish its goal of two-way communication and capacity building around contaminant issues. RCCs have been identified by the NCP as key people who have the capacity to bring the program closer to the North and also to bring community questions and concerns to NCP management. They have the expertise to do this because of their knowledge of the culture, people and geography of their regions. However, it is also important for RCCs to be familiar with the NCP. Until 1999, there had been little or no training for the position.

A training workshop for RCCs was planned to provide an opportunity for RCCs to get to know one another and to share ideas, experiences and knowledge. The workshop was also intended to cover many aspects of an RCC's responsibilities, including important logistical tasks such as time and budget management strategies and organizing workshops, background information about the NCP structure and its findings on contaminants in Phase I, evaluating communication priorities, developing educational materials and proposal writing and evaluation.

Activities in 1999-2000

The RCC Workshop was held September 27-28, 1999, in White Rock, BC to coincide with the NCP Results Workshop. The Results Workshop offered an opportunity for RCCs to learn more about NCP projects and to meet other NCP communicators and researchers.

RCCs attending the workshop were Norman Snowshoe from the Gwich'in Tribal Council, Lena Selamio from the Inuvialuit Regional Corporation, and Joanna Lampe from the Labrador Inuit Association. Lisa Nashak from the Nunavik Regional Board of Health and Social Services was scheduled to attend but due to flight delays, she did not reach White Rock until after the RCC workshop was finished. Nunavut RCCs were also invited but due to changes in leadership and restructuring in the newest territory, they had not been hired yet. The Métis Nation-Northwest Territories offered to provide them with assistance as well as materials provided at the RCC workshop once they were hired.

It was essential that participants in this session were comfortable asking questions, discussing issues and offering feedback. Although members of the NCP management committee asked to observe, it was felt that their presence would change the dynamics of the workshop. Instead, they were invited to attend the session in the afternoon of the second day to meet the RCCs. One member from the NCP Secretariat, Sarah Kalhok, was invited to attend the whole workshop and to participate in the discussions and activities. As the number of RCCs was small, this solution ensured that the key participants in the workshop, the RCCs, had a voice in workshop discussions as well as an opportunity to meet NCP management.

Results and Discussion

The RCC Workshop, held September 27-28, 1999, was well received. Participants welcomed the training session as they often feel as if they are working in isolation. Lena Selamio is responsible for the Inuvialuit communities of Holman, Paulatuk, Sachs Harbour, Inuvik, Aklavik and Tuktoyaktuk, where she works out of her own home. Norman Snowshoe looks after contaminant issues in the Gwich'in communities of Aklavik, Tsiigetchic, Inuvik and Fort McPherson. Joanna Lampe's region includes the Labrador coastal communities of Nain, Makkovik, Hopedale, Postville and Rigolet. Although her position is not officially that of an RCC, her organization is doing similar work and her contribution to the workshop was much appreciated. From workshop discussions it is clear that, for RCCs, NCP work is interesting and rewarding although there are some difficulties and frustrations.

The role of the RCC is to bridge the gap between community members and NCP scientists and management. Local concerns focus on local sources of contamination, but this does not fall within the NCP mandate. To build trust and respect, it is important to be able to address these non-NCP concerns. The RCCs feel that in order to communicate NCP information, they must be aware of the big picture in their region, in particular, who is responsible for specific local issues of water or waste management and what is being done about them. Therefore, in order to be effective, RCCs must be concerned with issues both within and without the NCP mandate.

It is important for southern management personnel to be aware of the unique challenges facing RCCs in the North. Some deadlines can be difficult to meet for logistical reasons. Travel is impractical at certain times of year, and lines of communication are not always reliable.

RCCs must be informed of any NCP activities in their regions, as it is likely that community concerns over these activities will be directed towards them. Information must be sent to the RCCs in succinct and well-organized packages. Their role is not to sort through and interpret volumes of information, but to "act as a conduit for two-way information exchange between the communities and the NCP." The RCCs felt that more training on long-range contaminants in the North is required, and that a course in chemistry would be useful.

Without exception, the RCCs felt that the job could easily take up more than the part-time position allows. In addition, it will be difficult to maintain continuity or attract the best people to the position, if other full-time jobs with higher salaries are available.

Conclusion and Recommendations

In their overall evaluation, RCCs felt that the workshop was enjoyable and interesting and that it provided much needed tools and information, although some issues remain unsolved. There was a lot of material to cover all at once. Some topics were "eye-openers" for some, therefore RCCs indicated that they would have benefited from a longer training course. They found it helpful to meet and share experiences with other RCCs, but expressed their hopes that more could have attended. It was also helpful to meet other people involved in the NCP, and RCCs appreciated being able to communicate to NCP management some of the difficulties they face in their positions. The need to be well informed of NCP activities was strongly expressed, as was the need for open communications with Aboriginal organizations. Some good discussion had to be cut short and planned activities cancelled because of the time constraints of the two-day workshop.

Several recommendations were made for future RCC training workshops. First, more time and/or workshops are needed to cover all of the material thoroughly. Second, the workshop leaders should consider holding the RCC training in conjunction with a Frontline Training course. Third, efforts should be made to ensure that RCCs are hired in time to receive training and orientation, and that the workshop is held at a time when it is reasonable to expect all RCCs have been hired. Fourth, more time and exercised should focus on how to develop proposals. Finally, experienced RCCs should be consulted in future development or modification of this workshop.

Discussions on communication priorities and methods of communication brought about some key points for RCCs and any other NCP personnel involved in communicating in the North to be aware of. One-on-one is the most effective (if not the most efficient!) means of communication in the North. Workshops are helpful, but all the information should not be presented at once; leave some information for follow-up workshops. Participants had several ideas for encouraging people to attend workshops, including promoting the workshop on the radio or in the paper, offering relevant draw prizes, and advertising well. Using local people in presentations, advertisements, posters and interviews draws attention to an event, as does using local people in organizing logistics and facilitating workshops. Furthermore, acknowledge local people for their assistance and participation.

Project Completion Date

Another workshop offering training and support for RCCs is planned for 2000-2001.

Country Food Nutrition and Health: Developing Effective Communication Strategies in Labrador (Year 2)

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Abstract

In the presence of growing information regarding the health risks imposed by contaminants and the social, cultural, economic and physical benefits provided by these foods, the task of developing and disseminating balanced messages in the many regions across the North is complex and challenging. Public health communicators in the North are faced with the challenge of developing messages that balance the information on both benefits and risks of traditional/country food diets and delivering them in ways that are accessible and understandable so that people can make informed decisions about their food use. Utilizing a literature review, key-informant interviews and focus groups, this study developed a list of regionally specific characteristics and guidelines for "effective" environmental health communications. Utilizing these guidelines, a communications strategy to convey results of local research on contaminants in country foods was developed, adapted and delivered in Makkovik and Nain, Labrador. Message reception through a number of commonly used pathways (poster, fact sheet, radio) and rough comprehension were assessed utilizing a telephone survey. Recommendations for effective environmental health communications in Labrador and a process for communication strategy development and assessment are made.

Objectives

Long-term

- To assist northern communicators and researchers in planning, constructing and disseminating messages and evaluating their effectiveness.

Short-term

1. To apply the knowledge gained from Year 1 of this project related to the characteristics of effective communications in Labrador to develop and test messages for dissemination.
2. To further adapt these characteristics with supporting empirical evidence from focus groups and a short community survey in two coastal communities.

3. To assess the effectiveness of a number of communication pathways regularly utilized for environment and health communications in Labrador.

Introduction

While our knowledge of Arctic contaminants and their effects has grown rapidly, the effective communication of this information to Northerners lags far behind. Research on Arctic contaminants, particularly over the last decade, has shed much light on modes of contaminant transport, distribution and deposition, as well as on biomagnification through food chains and contaminant exposure, toxicity and health effects on humans and ecosystems (Wheatley and Wheatley 1981, Barrie 1986, Barrie et al. 1992, Dewailly et al. 1994, Hild 1995). The Arctic is home to the Inuit of Canada, many of whom continue to rely on food from the sea and land for much of their subsistence. The presence of contaminants is widely perceived as a potential threat to the health and culture of Inuit. However, research and traditional knowledge have reported that traditional/country food is nutritionally, culturally, socially, and economically beneficial for people. Public health communicators are therefore faced with the challenge of developing messages that effectively balance the information on both benefits and risks of traditional/ country food diets, and deliver them in a way that is accessible and understandable so that people can make informed, healthy food and lifestyle choices in the North. Communication problems between researchers, public officials and community residents have been noted in the North in the past (Furgal 1999, Usher et al. 1995, Peters and Légare 1997). As a result of perceptions and misconceptions of risks, changes in traditional cultural practices have been seen in Inuit populations (Wheatley and Wheatley 1981, Dewailly et al. 1994). Choices made by individuals have lead to changes to traditional lifestyles resulting in negative impacts on personal health and social well-being (Wheatley and Wheatley 1981, Dewailly et al. 1994).

Despite efforts made by researchers, organizations and agencies to deliver accurate and easily understandable contaminants information, little work has been done to evaluate communications strategies used for health and environment issues (Usher et al. 1995, Furgal et al. 1995). Also, little has been done to develop regionally specific definitions and guidelines for "effective" communications on these issues. In Year 1 of this project, we used lessons learned from practice and research in environmental health risk communications outside Labrador, as well as knowledge gained from experienced communicators in Labrador, to develop regionally specific characteristics for "effective" environment and health communications (Andersen et al. 1999). In the second year of the project, 1999-2000, we used these guidelines to develop messages, test their comprehension among different sub-groups of the Labrador Inuit population, adapt and then deliver them, and assess commonly used methods of delivery in two coastal communities.

Activities in 1999-2000

Five different environmental health communications on topics related to local research on contaminants were developed using four different forms of media (fact sheets, video, radio interview and poster). Message content included one of the following: i) results of contaminant monitoring in Arctic char and ringed seals along the Labrador coast (fact sheet based on results of Muir et al. 1999); ii) results of the Labrador Inuit Health Commission regional health survey on contaminants and country foods (fact sheet); iii) results of the Labrador Inuit Association (LIA) Environmental Health Survey (video); iv) results and information on Year 1 of this project on environmental health communications (radio interview); or v) an update on the clean-up of polychlorinated biphenyl (PCB) contaminated soil from LAB-2, the long-range radar site in northern Labrador (poster developed by Environmental Sciences Group, Kingston, Ontario). Wherever possible, existing messages that had been developed on similar principles to those expressed in Year 1 of this project were used or adapted. Message development was guided by the principles of effective communications for Labrador developed in the previous year of this project (Andersen et al. 1999).

Focus group discussions (Morgan 1988) were conducted with four randomly selected individuals from each of five target groups among the population of Nain, Labrador. Focus groups were held with one group of each of women, youth, hunters, elders, and one mixed group of individuals. Focus groups reviewed all communication materials and discussed their level of attraction, ease of comprehension, and general suggestions for their adaptation and improvement. Results were recorded and organized in tabular format grouping common responses to the topics discussed.

The results of research reporting levels of heavy metals and organochlorines in Arctic char and ringed seal tissues from along the Labrador coast (Muir et al. 1999) was selected as the theme upon which to develop and evaluate a communication strategy in two Labrador communities. A fact sheet within a research newsletter, a poster and a radio interview were produced to disseminate the research results. These messages were developed using the characteristics developed in Year 1 as well as the recent assessments of messages and media in the focus groups as guidelines. The messages were then released in Nain and Makkovik, Labrador, using identified, common methods for disseminating this information. This included the posting of the summary poster in various locations around the community, the distribution of a fact sheet on the issue in the LIA research newsletter containing basic benefit and risk information on contaminants and details regarding projects going on in the region, and an interview broadcast on the regional radio station. Using a quota sampling strategy, random samples of hunters, women, youth, elders and a group of individuals randomly selected from the previous four sub-groups were selected from the LIA membership list for each community (Nain and Makkovik). Quotas were based on population representation and time and human resources available to conduct the interviews (Babbie 1990; Table 1). One week following the release of the information and the airing of the radio interview, a short telephone survey was conducted with these individuals to assess reception of the message (did people hear or see it?), comprehension (did they understand it? what questions did they have regarding the issue?) perceptions of these issues (how did they feel about these issues? what concerns did they have?), and ways this information could be better communicated in Labrador communities. Answers were transcribed and grouped according to common responses to question (Tables 2 to 6).

Table 1. Quota sampling numbers for targeted groups in Nain and Makkovik, Labrador

Community and Quota Group	Total Population	Target for this study	Sampled
NAIN			
Elders	29	20% = 6	2
Hunters	139	10% = 14	14
Youth	149	5% = 8	8
Women (child-bearing age)	146	10% = 15	14
Random	645	2.5% = 15	15
TOTAL	**1108**	**58**	53
MAKKOVIK			
Elders	24	20% = 5	5
Hunters	76	10% = 8	6
Youth	33	5% = 2 (min 5)	5
Women (child-bearing age)	58	10% = 6	6
Random	202	5% = 10	10
TOTAL	**393**	**34**	**33**

Results

Focus groups

Focus group discussions resulted in a number of recommendations for improving environmental communications in general and guidelines specific to print media, video and radio. A synthesis of these recommendations and points to consider when developing communications is presented below.

General Guidelines for All Media (print, radio and video)

- Include contact information for further inquiries, feedback, etc.
- Provide an explanation or definition for technical terms in a plain language format (this applies to Inuktitut translated text as well).
- Answer the questions: "How will this affect the country food I eat ?" "What does this mean to me ?" "Should I be concerned ?"
- Include comparisons with other regions of the North where possible.
- Proof-read Inuktitut translations by more than one translator for verification.
- Consider variations of the Labrador Inuktitut dialect between communities or settlements of origin in translations.
- Include both risk and benefit information.
- Provide information in clear and concise plain language.

Guidelines for Print Media (posters, fact sheets, etc.)

- Posters for public should provide one main message, limited text.
- Use eye-catching headlines on posters and newsletters.
- Use local photographs and maps where appropriate and highlight the region/community; use colour if possible.
- Include cartoons if targeting youth and/or students .
- Use bullet-points or short sentences, which are easier to read and more eye-catching than paragraphs.
- Use question and answer format.
- Highlight the summary.
- Logos used by organization should be visible.
- Consider that not all segments of the population understand graphs, e.g. elders.
- If research results are presented for a specific area or community, explain why only this area was chosen .

- Detailed research results are best for scientific or open house meetings, not for posting in the community.
- Provide Inuktitut and English versions in the same document if possible.

Guidelines for Videos

- Involve local people in presenting information.
- Youth prefer this medium over mediums such as newsletters, fliers and radio announcements.
- Youth prefer "people" issues more than "environmental" issues.
- Footage should not take away from the message and should reflect the information being presented.
- Make copies available for people to borrow and view on their own time; promote their availability.
- Introduction should state what kind of information will be presented and a summary at the end of the program should highlight the information presented.
- Footage of the local environment and people is appealing; the more familiar it is, the easier it is to get message across.
- Announce the program in advance through a variety of formats so that people know what kind of information will be presented and when.
- Inform individuals or groups who could utilize the video for educational purposes after initial release, e.g. public school teachers, about availability .

Guidelines for Radio Newscasts or Briefs

- People listen to different radio stations, so all should be utilized to reach a large number of people:
  - OKalaKatiget Radio – regional
  - Local radio station – e.g. for Nain it is CITN Radio
  - CBC Radio, either from Goose Bay, Labrador; St. John's Newfoundland, or Iqaluit, Nunavut (Inuktitut)
- The timing of the broadcast determines how many people you reach – e.g. CBC Radio from Goose Bay reaches many people because it broadcasts early in the morning before people go to work, out wooding, etc. and they listen to it for weather, school closures, etc., and is the only radio station on at that time for the region (preferred time slots: morning show from 6:00-9:00 am, noon hour, afternoon and evening).
- Radio is not effective for targeting youth.
- Local project staff should present the information.
- Scripted interviews or reading is viewed as monotone or boring.
- Give background information on organizations involved that are based outside of the region.
- Use format appropriate for the information being presented, for example a news clip instead of an interview when informing public about an ongoing project.
- Provide examples to illustrate the point being made.

Survey results

A total of 86 individuals representing hunters, women, youth, elders and a group representing a randomly comprised group of individuals from each community were interviewed one week following the release of the communications in Nain and Makkovik, Labrador. Fewer than half of the people interviewed in each community recalled seeing or hearing information on environment and health issues, contaminants in the environment, etc., in the past week (Table 2). In Nain, the most commonly reported test messages seen were the posters in the post office and health clinic and the notice on the cable television channel notice board (roll-up) informing people that newsletters were available in the post office on this issue (Table 2). In Makkovik, the most commonly reported item seen was the poster in post office and the Markland Building (common building in town housing health offices, stores, etc.) (Table 2). Among individuals reporting to have seen or heard this information, more than half in each community, reported that the general message related to contaminants, food safety or pollution. Two individuals in Nain reported that the main message seen or heard related to communication issues, or parasites in wildlife (Table 3). In response to questions regarding their behaviour relating to communication reception pathways, survey participants indicated that the majority had been to the post office within the past week (Nain = 35/53, Makkovik = 24/33) and many visited the post office three or more times per week on average (Table 4). Less than half of Nain respondents had listened to the regional radio broadcast in the last week, and approximately the same number reported being regular (three to four times per week) listeners to this source of media. In Makkovik, approximately half of all respondents reported listening to the regional radio broadcast sometime in the past week and reported being regular listeners (Table 4). More than 75% of respondents in both communities reported regularly picking up newsletters when they were available in the post office in their community (Nain = 44/53, Makkovik = 21/33) (Table 4).

The large majority of individuals interviewed in each community and from each sub-group reported that they felt country foods were safe to eat (Table 5). Those that

Table 2. Recollection of communicated information in Nain and Makkovik

Group	N	Have you seen or heard information released by the LIA Research Office recently?		What did you see or hear?												
		Yes	No	A	B	C	D	E	F	G	H	I	J	K	L	M
NAIN																
Random	15	7	8	1	2	1	2	0	0	2	0	0	0	0	4	1
Hunters	14	7	7	0	0	0	2	0	0	1	0	0	0	0	3	2
Women	14	5	9	0	3	0	2	0	0	0	0	0	0	0	4	0
Youth	8	3	5	0	2	0	0	0	0	0	0	0	0	0	0	2
Elders	2	1	1	0	1	0	0	0	0	0	0	0	0	0	1	0
TOTAL	**53**	**23**	**30**	**1**	**8**	**1**	**6**	**0**	**0**	**3**	**0**	**0**	**0**	**0**	**12**	**5**
MAKKOVIK																
Random	10	1	9	0	0	0	0	0	1	0	0	0	0	0	0	0
Hunters	6	2	4	0	0	0	0	0	2	0	0	0	0	1	0	0
Women	7	3	4	2	0	0	0	0	2	0	0	0	0	2	0	0
Youth	5	0	5	0	0	0	0	0	0	0	0	0	0	0	0	0
Elders	5	2	3	1	0	0	0	0	2	0	0	0	0	0	0	0
TOTAL	**33**	**8**	**25**	**3**	**0**	**0**	**0**	**0**	**7**	**0**	**0**	**0**	**0**	**3**	**0**	**0**
GRAND TOTAL	**86**	**31**	**55**	**4**	**8**	**1**	**6**	**0**	**7**	**3**	**0**	**0**	**0**	**3**	**12**	**5**

A = OK Radio broadcast
B = Post in post office
C = Poster in school
D = Poster in health clinic
E = Poster in LIA Research Office
F = Poster in Markland Building (Makkovik)
G = Newsletter in postoOffice
H = Newsletter in school
I = Newsletter in health clinic
K = Newsletter in Markland Building (Makkovik)
J = Newsletter in LIA Research Office
L = TV roll-up ad about newsletter (Nain)
M = Other

Non-responses are responsible for some groupings of answers not totaling N.

Table 3. Recollection of message content in Nain and Makkovik

Group	N reporting seeing or hearing material	Do you remember the general content of the message?						
		Yes	No	Contaminants	Communication	Food Safety	Pollution	Parasites
NAIN								
Random	7	5	2	2	1	1	0	0
Hunters	7	7	0	3	1	2	0	2
Women	5	0	5	0	0	0	0	0
Youth	3	2	1	2	0	0	0	0
Elders	1	1	0	1	0	0	0	0
TOTAL	**23**	**15**	**8**	**8**	**2**	**3**	**0**	**2**
MAKKOVIK								
Random	1	1	0	1	0	0	0	0
Hunters	2	2	0	0	0	1	2	0
Women	3	2	1	1	0	0	1	0
Youth	0	0	0	0	0	0	0	0
Elders	2	2	0	2	0	0	0	0
TOTAL	**8**	**7**	**1**	**4**	**0**	**1**	**3**	**2**
GRAND TOTAL	**31**	**22**	**9**	**12**	**2**	**4**	**3**	**2**

Non-responses are responsible for some groupings of answers not totaling N.

Table 4. Communication reception behaviour in Nain and Makkovik

		Have you been to the post office this past week?		How often do you visit the post office/week?			Have you listened to OK radio in the last week?		Do you listen to OK radio regularly (3-4 times/week)?			Do you take newsletters from the post office when they are available?	
Group	**N**	**Yes**	**No**	**< 3**	**3-5**	**> 5**	**Yes**	**No**	**Yes**	**No**	**Sometimes**	**Yes**	**No**
NAIN													
Random	15	9	5	8	2	4	5	9	5	8	1	11	3
Hunters	14	10	4	8	3	3	6	8	5	9	0	14	0
Women	14	10	4	3	6	5	6	8	6	8	0	10	4
Youth	8	5	3	5	2	1	3	5	5	3	0	7	1
Elders	2	1	0	0	0	1	0	1	0	1	0	2	0
TOTAL	**53**	**35**	**16**	**24**	**13**	**14**	**20**	**31**	**21**	**28**	**1**	**44**	**8**
MAKKOVIK													
Random	10	7	2	4	5	1	6	4	4	4	2	7	3
Hunters	6	3	3	3	3	0	4	2	4	2	0	5	1
Women	7	7	0	1	1	0	3	4	5	2	0	3	4
Youth	5	5	0	2	2	1	0	5	1	4	0	2	3
Elders	5	2	3	5	5	0	5	0	3	0	2	4	1
TOTAL	**33**	**24**	**8**	**15**	**15**	**2**	**18**	**15**	**17**	**12**	**4**	**21**	**12**
GRAND TOTAL	**86**	**59**	**24**	**39**	**39**	**16**	**38**	**46**	**38**	**40**	**5**	**65**	**20**

Non-responses are responsible for some groupings of answers not totaling N.

felt they were not safe to consume indicated concerns for contaminants, parasites or pollution from military sites or town dumps as the most common reasons for their response (Table 5). Despite the reported trust in the safety of country foods, many individuals still reported some concerns regarding country foods in general (Table 5). These included such things as, the levels of contaminants in wildlife, the impacts these contaminants have on the health of species, specific concerns regarding the environmental impacts of activities related to the Voisey's Bay mining project, food safety issues such as diseases in animals, and a lack of information being released to the public on these issues.

When asked to make suggestions to improve the way this information is communicated in their community, individuals reported a number of pathways and forms of media, some commonly used and others not regularly exploited in communication strategies (Table 6). A common request for more information in various forms (posters and newsletters) available around town was reported by individuals in both communities, while a large number of individuals from Makkovik specifically suggested home delivery of newsletters or personal home visits to communicate this information. All youth interviewed in Makkovik suggested distribution of information in the school (Table 6).

Discussion

The guidelines synthesized in this project are not new to the field of risk communication or environmental health communication. Many of the points mentioned here exist in the literature reviewed in the first year of this project (see Andersen et al. 1999) and as recommendations from other studies (e.g. Usher et al. 1995, Furgal et al. 1995, Furgal 1999). However, through this project they have been adapted to better reflect the experience, thoughts and perspectives of communicators and individuals in Labrador communities. Some of these are applicable to other regions and communities, however, others are perhaps specific to Labrador. For example, even beyond the simple translation of technical terminology into plain English and Inuktitut, the impact of different community-of-origin dialects of Inuktitut on reading comprehension of printed material can be significant, as reported in focus group discussions. Further, the difficulties in interpreting graphically presented information by many focus group participants is something that must be considered in

Table 5. Perspectives on country foods from survey participants in Nain and Makkovik

		Is country food safe to eat?			If no, why not?			Do you have concerns about the country foods you consume?	
Group	**N**	**Yes**	**No**	**It depends**	**Contaminants**	**Pollution from military sites and dumps**	**Parasites**	**Yes**	**No**
NAIN									
Random	15	12	1	2	1	0	0	1	0
Hunters	14	12	2	0	1	1	0	2	4
Women	14	13	1	0	0	0	1	0	5
Youth	8	8	0	0	0	0	0	1	0
Elders	2	0	1	1	2	0	0	1	0
TOTAL	**53**	**45**	**5**	**3**	**4**	**1**	**1**	**5**	**9**
MAKKOVIK									
Random	10	10	0	0	0	0	0	4	5
Hunters	6	6	0	0	0	0	0	5	1
Women	7	6	0	1	1	0	0	6	1
Youth	5	5	0	0	0	0	0	4	1
Elders	5	5	0	0	0	0	0	4	1
TOTAL	**33**	**32**	**0**	**1**	**1**	**0**	**0**	**23**	**9**
GRAND TOTAL	**86**	**77**	**5**	**4**	**5**	**1**	**1**	**28**	**18**

Non-responses are responsible for some groupings of answers not totaling N.

communicating this type of information in Labrador and other northern regions.

Many of the recommendations concur with earlier work regarding the need for involvement of local people as the first communicators of this information, the need to utilize multiple media and pathways to deliver messages to a wider audience, and the need to deliver both risk and benefit information on these topics (Andersen et al. 1999, Furgal 1999). Despite this, many of the points raised here are not commonly followed in current communications in the region for various reasons (Furgal 1999). Financial and time constraints often limit preparation, review and adaptation opportunities for communication materials and strategies on many issues. However, it is important to note that the degree of adaptation in message content, wording, and presentation resulting from the recommendations made by focus group participants here was substantial and is an indication of the level of effort required to develop communications that effectively reach local people*.

Fewer than half of all people interviewed in both communities reported seeing or hearing the released test information yet more than half reported having been in places where material was posted or available, or were listening to radio programs broadcasting the information for this study (Tables 2, 4). Seasonality affecting when individuals are in the community, community events, multiple messages on similar issues, local and regional contemporary issues as well as day-to-day personal concerns all compete for individuals' attention in public communication forums in northern communities today. Many individuals still rely on, and as seen in this survey, request face-to-face contact for reliable communication on important community issues; however, the number of pathways and sources of media and messages individuals are exposed to everyday continues to increase in the North, as elsewhere. Many of those in Nain reporting seeing the information available reported knowledge of it through seeing the notice on the local cable television notice board (roll-up). Many traditional forms of communication often relied upon in the North, such as community meetings and radio, were not suggested as often as others as means of delivering information. Instead, many individuals suggested delivering messages door-to-door in written or verbal form, or through increased regional

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* Due to space limitations it was not possible to include copies of tested material in full. Copies of this material can be obtained by contacting the LIA Research Office in Nain, Labrador.

Table 6. Suggestions to improve communications provided by survey participants in Nain and Makkovik

		Do you have any suggestions to improve communications?								
Group	**N**	**Yes**	**No**	**More newsletters / posters around town**	**Home delivery/ visits**	**P.O. Box delivery of material**	**More use of radio**	**More use of TV roll-up**	**Hold community meetings**	**Post info in the school**
NAIN										
Random	15	7	6	4	2	0	1	1	0	0
Hunters	14	7	7	3	2	1	2	0	0	0
Women	14	11	3	3	2	4	2	1	0	0
Youth	8	5	3	2	1	0	1	0	1	0
Elders	2	1	1	0	1	0	0	1	0	0
TOTAL	**53**	**31**	**20**	**12**	**8**	**5**	**6**	**3**	**1**	**0**
MAKKOVIK										
Random	10	10	0	5	5	0	2	1	0	0
Hunters	6	6	0	2	5	0	2	0	1	0
Women	7	5	2	4	3	0	2	0	1	0
Youth	5	5	0	0	0	0	1	0	0	5
Elders	5	5	0	3	3	0	2	0	0	0
TOTAL	**33**	**31**	**2**	**14**	**16**	**0**	**9**	**0**	**2**	**5**
GRAND TOTAL	**86**	**62**	**22**	**26**	**24**	**5**	**15**	**4**	**3**	**5**

Non-responses are responsible for some groupings of answers not totaling N.

and local television and radio programming. Some of these newer (e.g. local cable television) forms of media available in some communities need to be assessed for their feasibility in delivering messages to a wide range of audiences as they are not currently exploited, yet are increasing in their importance as local information sources in many communities.

Although most individuals in both communities reported picking up newsletters when available in public locations, few reported seeing the newsletters in this study. The visibility and daily availability of the newsletters are critical factors in interpreting this observation. Some individuals reported not looking at postings on the wall or picking up newsletters while in the post office because of a lack of interest or time. Difficulties have been noted in the past with newsletters and mail-outs being quickly discarded (wasted) in the post office in Nain and thus we took a conservative approach to the number available for pick up in public locations (15 to 20 available all the time) and attempted to ensure they were always available by frequent personal checks to these locations. It is possible that newsletters were not visible enough in these locations, were quickly depleted for one of a number of reasons, or that a one-week release period is too short upon which to base this evaluation. The one-week viewing period during which this information was released was short considering that a number of individuals reported only visiting the post office fewer than three times per week. The locations where posters and newsletters were placed are reported to be frequently visited by most groups, with the exception of elders, in both communities and should continue to be used to release this information.

One challenge highlighted by this survey is the difficulty in reaching youth with many of these messages. Although they visit the locations tested here and are often exposed to the other forms of media discussed, few reported seeing or hearing this information. Many reported a lack of interest in these issues during Year 1 of this study (Andersen et al. 1999). As these are important issues for future individual and family decision makers, it is important that they are communicated to and understood by this segment of the population. Therefore, further work is required in collaboration with youth to develop methods and tools to better communicate this information to them in Labrador. It should be noted here that the Internet, a popular media considered for communication with youth in other regions of the North and elsewhere, was not evaluated in this study, as access is currently limited in Labrador communities. However, this situation is changing with the placement of a regional access Internet "node" (eliminating the need for a long distance hook up to a server) and future communications with youth should consider evaluating this medium.

Among those reporting having seen the message, hunters were most likely to remember the content (Table 3). Hunters and women of child-bearing age have perhaps been most exposed to this information in the North through past communication efforts relating to food safety, nutrition and wildlife concerns conducted under this (Northern Contaminants Program) and other northern programs. These individuals are most often involved in collecting, distributing and preparing the food for consumption; however, there is a need to reach others in the community with this information as all are individual decision makers and youth are the family decision makers of tomorrow.

It must be noted that this survey did not assess comprehension in any detail and lends itself to an indication of main message recollection alone. The attention that contaminants in traditional/country foods have received in many regions in the North in the recent past may have significant influence on the results here. Despite this, from a public health perspective it is reassuring to know that the majority of individuals in Nain and Makkovik, Labrador, express a balanced understanding of contaminants in country foods. The large majority report the belief that country food is safe to eat today, with few expressing uncertainty. However, local authorities should note that this does not exclude the fact that many have numerous concerns regarding the country foods they consume, including such things as their level of contaminants, trends in these levels, their effects on the wildlife and the safety of these animals to eat.

Conclusions

This study represents a beginning in structured communication evaluations on contaminants and country food issues in Labrador. Through focus group discussions, many communication guidelines were adapted and expanded for Labrador communicators. The needs to consider different local Inuktitut dialects, to develop different illustrative ways to convey messages, and to conduct some form of pre-testing of messages before wide distribution were highlighted here. The survey illustrated some new pathways that require further exploitation in order to reach diverse groups in Labrador communities, and the demand for home-delivered information in written or verbal form. Additionally, it showed the need to further explore and develop ways to communicate with youth on many of these issues. The development of the first edition of a research newsletter may help to increase capacity to communicate this type of information in the region by providing a tested template. The study demonstrated the need to plan, utilize, and assess a variety of methods and media for communicating with specific target groups within northern audiences. It outlines a simple strategy for communications planning, development and evaluation and supports the argument for evaluation exercises. As well, it is suggested that standard formats and locations be identified through evaluations and continuously used for regular communications of this type. Finally, implications for researchers communicating results in Labrador and other northern regions are far reaching. More directed thought, time and local support is needed to effectively deliver research results and general messages on these issues which affect the daily lives of people living in the North.

Date of Completion

This project is now complete. However, community reports and publications of results and their implications for community communications on environmental health issues will continue to be produced throughout 2000 in cooperation with LIA and regional communicators.

Acknowledgments

The researchers would like to express their thanks to all who participated in interviews, focus groups and responded to the survey as part of this project. Without their interest and cooperation, this project would not have been possible. Additionally, the assistance of Carol Gear was invaluable in carrying out the survey in Makkovik, Labrador, and for this we thank her.

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Design and Development of an Inuit Tapirisat of Canada Website and CD-ROM Communication Package on Northern Contaminants

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Abstract

The aim of this project was to develop a website and CD-ROM to communicate information on contaminants and the Northern Contaminants Program (NCP) to Inuit and other interested individuals. This website (www.tapirisat.ca) enables Inuit to make informed decisions regarding the use of traditional/country food, provides access to information on contaminant research both past and present, and establishes a framework for Inuit participation in the NCP. The website was designed to provide a forum for Inuit knowledge and ideas to be shared with a southern audience, as and to make this information available to a much broader audience internationally. The use of new media will not replace the importance of face-to-face exchange of information through meetings and workshops; instead, it introduces a vital tool for Inuit to expand the communication capabilities with Inuit Tapirisat of Canada and the NCP.

Objectives

1. To work with the regional contaminant coordinators (RCCs) and the territorial contaminants committees to determine core objectives of a communication package on contaminants and to design and develop a web-based product for reaching these objectives.
2. To develop linkages between the Inuit Tapirisat of Canada (ITC) Indigenous knowledge database and the information collected as part of the 1975 ITC Land Use and Occupancy Project.

3. To develop a "library" and/or storage facility, which will contain information that has been collected from Northern Contaminants Program (NCP) and ITC publications, workshops, meetings and personal communication.
4. To communicate general and technical information about northern contaminants to both northern residents and non-northern audiences, who seek to learn about contaminants in the north, their sources and impacts.
5. To provide much needed information in a format that can be easily updated in order to provide the most current information and research data.

Introduction

Several NCP studies have been completed and a large body of data on contaminants in the environment, wildlife and humans, as well as on Northern diets and nutrition has been collected, with preliminary or final results appearing in a variety of documents. As well, a significant amount of information on topics related to contaminants has resulted from meetings, workshops, conference calls, phone-in shows, public service announcements, videos and frontline training. In spite of this vast amount of information, there is still a lack of understanding among Inuit communities about contaminants. Our intention with this project is to provide a venue where this information is more easily accessible.

The purpose of this project is to undertake a new approach for communicating research data and information about contaminants in the North while melding the perspectives of both Inuit and scientists. The availability of a website and CD-ROM yields new communication options with the flexibility to incorporate relevant information from various sources into a comprehensive communications package. The use of multi-media will not, by itself, resolve the difficulties of communicating about contaminants. There will most likely always be difficulties using the Internet in the Arctic due to inaccessibility. Certainly the use of the Internet and CD-ROM technology does not diminish the importance of the continuing use of face-to-face exchange of information through workshops and meetings or the more formal documentation of information through written reports. Rather, it builds upon these.

The need for information on contaminants is essential with respect to the harvesting of traditional/country foods. In the last decade, the risks associated with harvesting of traditional/country foods across the Arctic has become an increasingly important issue for Inuit. In order to address these concerns, Inuit, as consumers of country foods, must have access to relevant and meaningful information. Such information empowers them to make informed decisions about the harvesting and consumption of these foods. A website that can post new material as soon as it becomes available has the advantage of maintaining an up-to-date bank of information about contaminants.

The primary audience of the ITC/NCP website and CD-ROM communication and education package are the consumers of country foods. At the same time, however, the package will be available to the population at large in both the North and the South as it is important that they have a more in-depth understanding of the contaminant issues and concerns that effect Inuit and other Northerners.

This website and CD-ROM will therefore provide a very powerful educational tool to our northern target audience while also serving as an educational tool for other audiences. Since the potential users of the ITC website are worldwide, this communication package will be extremely valuable in presenting the global sources and the global responsibilities for northern contaminants. It is critical to highlight transboundary issues as they relate to the circumpolar area.

Activities in 1999-2000

Discussions on the primary objectives and audience for the website and the selection of information that should then be included were held in various community meetings and gatherings. A listing of topics and a "paper site" that identified the organization and interactivity of the website and CD-ROM were developed. The paper site also illustrated the primary icons and other design features of the website.

Individuals from the NCP and other organizations were contacted to obtain the most recent information on contaminants and visual material such as charts and graphs. Maps and other visual materials that illustrate key findings about contaminants were prepared. Finally, programming of the design of the website, as well as its on-screen structure and navigation, was carried out. The website will be burned onto CD-ROM and distributed to those communities without Internet connections.

Results

The activities described above have resulted in the development of a functioning prototype of a website and CD-ROM based communication package on northern contaminants. The site uses a series of maps to convey information about contaminant sources. Charts and diagrams are used to communicate more specific information about contaminants and their potential impacts on Inuit. The interactivity allows a user to select specific topics of interest or choose a particular way to navigate the site.

The site will now be integrated into the ITC website (www.tapirisat.ca) as a stand alone unit of the larger site. The feedback from users will be recorded, which will allow any problems to be traced and the strengths to be highlighted in order to maximize the use of the Internet for communicating about contaminants.

Discussion

The necessity for communicating information about contaminants and their potential impact has been an underlying concern in phase I and II of the NCP. Contaminants issues are complex, the data is not always easy to explain, and the interpretation of data in terms of its use for risk assessment and intervention does not always provide clear answers to consumers of country foods who are concerned about the safety of their food sources. The ITC website will be used as a means for communicating information about contaminants but cannot provide answers itself, it is simply another method of communicating information. One of the objectives of this project is to see if these electronic options can actually become effective and advantageous for communicating about contaminants.

One potential advantage yet to be explored is the use of the website to provide information to special groups, such as the soon-to-be-appointed Regional Contaminants Coordinators (RCCs). The expectation is that the website will be a very useful tool for sending information to the RCCs and encouraging the RCCs to talk amongst themselves, based on the information derived from the website. At another level, the website will be used to reach a broader audience comprised of individuals, educational institutions, regional and community organizations, and scientists conducting research within Inuit regions.

This contaminants website and CD-ROM project is linked to the traditional Inuit knowledge database project. Information from both projects will be displayed together on the ITC website as part of an approach to achieve a more in-depth understanding about contaminants in the North.

Expected Project Completion Date:

The website and CD-ROM will be posted at the end of July 2000.

Acknowledgments

ITC would like to thank the individuals from the NCP who have been very helpful in providing information and answering questions related to the best use of this information.

Design and Development of a Prototype for an Indigenous Knowledge Database for Integration with the Northern Contaminants Program

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Abstract

Inuit possess a detailed and profound knowledge of their environment. Canadian Inuit have made it clear that their traditional knowledge, or *Inuit Qaujimajatuqangit*, can be valuable for enhancing and ensuring a clearer understanding of the issue of contaminants in the circumpolar Arctic; but to enable this to happen, Inuit knowledge must be organized and presented in a forum that is understandable, rigorous, and readily available to specific as well as broad audiences. The intention of this project was to use the earliest available Inuit knowledge data set (i.e. from the 1975 Inuit Tapirisat of Canada Land Use and Occupancy Project) to develop a prototype Inuit knowledge database that contains information from Inuit living in five communities located on southern Baffin Island: Kinngait, Kimmirut, Iqaluit, Pangnirtuuq and Qikiqtarjuaq. This information was collected at a time when Elders' memories and knowledge dated back to the significantly distant past. Hence, this database provides a solid foundation for temporal trend knowledge and knowledge of the land use patterns of today. This prototype provides a forum for Inuit to participate more fully in contaminant issues and in the Northern Contaminants Program (NCP) , and will also inform scientists of traditional knowledge information that they often have difficulties in locating and assessing. The use of Inuit traditional knowledge combined with the "western science" of other NCP researchers maximizes our understanding of the environment and ecosystems of this vast Arctic region.

Objectives

1. To establish an Inuit knowledge database prototype about the environment and land use using available information collected as part of the 1975 Inuit Tapirisat of Canada Land Use and Occupancy Project.
2. To organize and present Inuit knowledge of land use in a forum that all Inuit communities can access.
3. To develop this prototype in a format that makes the information readily available to a broad audience

through the internet, using the Inuit Tapirisat of Canada (ITC) website (www.tapirisat.ca) and by an interactive CD-ROM.

4. To conduct an evaluation of the completed prototype with input from Inuit regional representatives, the Regional Contaminant Coordinators (RCCs) and other potential users such as Northern Contaminants Program (NCP) staff, scientists and other educators, in terms of the capacity to enhance communication and understanding among and between Inuit and the NCP.

Introduction

> *There are many ways to be poor in today's world, not having the right kind of information represents a certain kind of poverty. As long as outsiders decide what is important and are in a position to ask all of the questions, we will never be in able to solve our own problems.*

This statement on the importance of information was made by the late Mark R. Gordon who clearly understood the role that information must play as a vital ally in helping Inuit of Canada to meet challenges and take advantage of the many opportunities that will define the next stages of Inuit social, economic and political development.

In Arctic research, the knowledge of the Inuit is a most neglected consideration. Inuit possess a detailed and profound knowledge of their environment, which has ensured their survival and self-sufficiency over countless generations. Today, this unique knowledge is just as important as it always has been to ensure the proper, efficient, safe and sustainable use of renewable resources. Canadian Inuit have made it clear that their traditional knowledge is also valuable for enhancing and ensuring a more clear understanding of the problems of contaminants in the circumpolar Arctic. Traditional knowledge can play an important role in our understanding of areas such as human health, wildlife monitoring and culturally appropriate methods of communication and education on contaminant issues. In order for Inuit traditional knowledge to be incorporated into programs such as the NCP, it must first be organized and presented in a forum that is understandable, rigorous, and readily available to specific as well as broad audiences.

Inuit Knowledge, or *Inuit Qaujimajatuqangit* has been identified by the NCP as an essential source of information that can be used in combination with other types of information (e.g. scientific data) for addressing a wide range of contaminant and related issues. This unique knowledge provides information that is essential when working with Inuit on the management of risks posed by contaminants and the maintenance of a traditional diet. In turn, decisions surrounding the benefits and risks of consuming traditional diets influence Inuit traditional harvesting patterns and lifestyles.

Inuit know the importance and potential of their knowledge. They need to ensure that communities and organizations use the full potential of this knowledge to understand contaminants and their impacts. At the same time, Inuit are concerned that scientists and policy makers involved with northern contaminants may not share this perspective, primarily because they have not had an opportunity to work with this type of information. If there is to be a consensus within the NCP about what constitutes Inuit knowledge and how it can be collected, processed and incorporated into the scientific research, communication and education mandates of the NCP, then researchers and policy makers need to be exposed to how Inuit knowledge can be useful and necessary in addressing questions of contaminants.

Under the NCP, researchers are encouraged to respect and include Inuit knowledge in their own research programs. Experience has shown, however, that the very nature of Inuit knowledge makes it difficult for outside researchers to do so. There are many reasons for this, but two are of particular interest. First, the collection of Inuit knowledge requires specialized expertise and a specific methodology. It cannot be systematically collected simply as a by-product of other research goals and methodologies. Second, Inuit feel strongly that the collection, interpretation and communication of their knowledge must be under Inuit control and supervision. Inuit are simply seeking an effective participatory role in the research process. The development of this Inuit traditional knowledge database prototype is a step in that direction.

There has long been the question of how to create an information tool based on traditional knowledge. Determining how to organize and present traditional knowledge is a real challenge. ITC felt that the place to begin was to use information already available through the 1974 Inuit Land Use and Occupancy Project (Freeman 1976) to establish a well-organized and user-friendly database of this environmental knowledge. The attraction in using this existing information is that the information was collected with the help and knowledge of participating hunters, especially the elders at that time. This represents a source of information that is irreplaceable as part of the knowledge base of Inuit culture. The information was collected at a time when elders' memories and knowledge dated back to the significantly distant past. Once this information is made readily available, it will provide a critical link between two knowledge sources:

that of present day hunters and that of the hunters of the 1970s.

This database prototype utilizes maps as the primary way to express both Inuit knowledge and land use information and therefore the database is organized within the framework of a Geographic Information System (GIS). The interface developed for the website presentation allows for accessing the information on both a community and regional basis.

This database plays an important role in addressing the concerns Inuit have about the importance of including their own knowledge as part of the scientific communication and educational mandates of the NCP. It provides a foundation of ecological knowledge necessary for Inuit to participate with issues surrounding contaminants.

Activities in 1999-2000

The five activities that were undertaken to complete this project are described below.

Activity 1

This involved locating and assembling all of the relevant information on land use and Inuit ecological and environmental knowledge required for the pilot communities located on southern Baffin Island (Kinngait, Kimmirut, Iqaluit, Pangnirtuuq and Qikiqtarjuaq) and surrounding areas. This information included items such as unpublished field notes and field maps that were collected during the ITC Land Use and Occupancy Project (Freeman 1976). All notes were photocopied and, when possible, the typed copies, though old and not in good condition, were scanned to create an electronic copy.

Activity 2

This involved the creation of electronic copies of the field maps. The land use maps were created by digitizing the land use patterns based on the original field maps and the maps published in the atlas volume (Volume 3) of the Inuit Land Use and Occupancy Report (Freeman 1976). The ecological maps were created by digitizing information on selected species from the field maps, but primarily from using the geographic references linked to written ecological descriptions in the field notes to create a new set of ecological maps for the selected species.

Activity 3

This involved developing a series of base maps that met the technical specifications for downloading onto the web. The electronic maps were created using the Digital Map of Canada (US Defense Mapping Agency 1994) and the Digital Map of the World (Environmental Systems Research Institute 1992). These maps were then scaled to the needs of the web. A 15 cm x 15 cm format was selected. At the same time, a larger format was created for a set of paper maps. The final stage of work was to establish the design criteria for the maps and to create an electronic atlas. The digitized information on major species harvested by Inuit was placed on the maps along with supporting written text in both English and Inuktitut, which provides in-depth detail on seasonal land use of specific areas, resources use and the economic, social and spiritual importance of areas to Inuit.

Activity 4

This included designing all of the interfaces that are required to search for and display this information on the web. This involved a series of reasonably complex activities and required a concise blueprint for organizing and establishing linkages between the different types of on-screen information. The next step was to write the programs needed for accessing the information. The final step is to carry out the testing and correcting of the interface and the information that is retrieved through the interface and to then integrate this interface into the larger ITC website.

Activity 5

This involved the evaluation stage. Review prototype with regional Inuit organizations, RCCs and NCP managers and scientist to discuss implementation and future use of the prototype. This will be completed when the RCCs are hired.

Results

The activities described above have resulted in the development of a functioning prototype of a web-based atlas and descriptive text on the content of Inuit knowledge for the selected pilot communities. This information can be accessed at www.tapirisat.ca.

Discussion

The completed prototype is intended to test the possibilities for enhancing the understanding and the use of Inuit environmental knowledge and land use information and, by so doing, to help improve the use of this information within the NCP. Providing a common and easily accessible database in the form of maps and descriptive text can guide researchers to very detailed site-specific information about what Inuit know about that area ecologically. This website is designed to provide researchers with a place to turn to in order to obtain the level of information which they may require to get a more complete picture of resource activity in their area of study.

The use of the ITC website makes this information widely available, but at the same time it has certain limitations in terms of the map size and allowable complexity of the information that can be placed on these maps. The database, however, is adaptable and can accommodate the production of more traditional paper maps.

Since it is certain that new technology and software will soon bring drastic improvements to the movement of GIS type information over the World Wide Web, this project should be viewed as positioning ITC and the NCP with an initial foundation for advancing the use of this technology for communication and scientific purposes. This database will be added to and updated continually. It is expected that when new information comes in from the NCP that it will be incorporated within the site.

This database will enable Inuit knowledge and expertise of the Arctic environment to be used for planning, execution and analysis of research, leading to better results for everyone concerned. The time has come when Inuit feel that their knowledge of the environment should be used for research so that this information can be used to make more balanced decisions. The development of this traditional knowledge "atlas" therefore is an ideal way to classify Inuit knowledge for both our own and others' use as a reference for research or for cultural fulfilment.

The expectation is that this database will be added to on as needed. It is designed to incorporate feedback and so that future development can occur, such as adding additional Inuit communities and data, adjusting data, and adding NCP findings and other information as they become available. The development of this traditional knowledge atlas will allow researchers a chance to quickly get "up to speed" on Inuit land use, traditional ecological knowledge and resource management while at the same time including and supporting Inuit opportunities to get into the scientific community without totally giving up their own ways and ideas. This database will provide an opportunity for Inuit to produce the data and information that will comprise our knowledge as Inuit.

Expected Project Completion Date

The prototype is being tested and will be posted in the summer of 2000.

Acknowledgments

ITC would like to thank the Inuit elders and hunters of the 1970s for the gift of knowledge they provide to the Inuit of today.

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Gwich'in Settlement Area Regional Contaminants Coordinator

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Abstract

The Gwich'in rely heavily on traditional foods as critical components of their diet. Due to concerns in Gwich'in communities regarding contaminants in the food chain, a Regional Contaminants Coordinator (RCC) position was established. This position was developed so that the Gwich'in would be able to participate in the Northern Contaminants Program (NCP) and so that communities had someone in the Gwich'in Settlement Area to turn to with questions and concerns. During 1999-2000, the RCC developed and delivered programs to address those concerns of the Gwich'in. One major concern was the lack of communication with respect to information from past and present scientific studies conducted in the region. The RCC has begun to address those concerns by holding public information sessions in the communities that comprise the Gwich'in Settlement Area. Resource persons from the NCP and affiliated organizations participated in the workshops. The RCC attended local meetings in each of the communities to address further concerns and to pass on information related to contaminant concerns. Furthermore, the RCC developed programs that would address specific concerns under the NCP blueprint for Local Contaminants Concerns. In this year, it was the collection and analysis of fish from Fort McPherson, Northwest Territories. Finally, the RCC participated in NCP activities including the RCC Training Workshop, the NCP Results Workshop and the Northwest Territories Environmental Contaminants Committee meetings.

Objectives

1. To promote the role of the Gwich'in Tribal Council as a partner in the Northern Contaminants Program (NCP).
2. To assist Gwich'in communities to identify and develop research programs to address contaminant concerns and local issues.
3. To inform and educate the public about contaminants within the Inuvik region.
4. To present the results from studies that are relevant to the consumption of traditional foods in the Gwich'in Settlement Area (GSA).
5. To increase capacity at the regional/local level.
6. To coordinate regional studies.
7. To identify complementary environment issues and funding sources.

Introduction

The Regional Contaminants Coordinator (RCC) position has been identified as an integral part of the NCP. These frontline workers are important in the communications aspect of the NCP. Until now, the Gwich'in were not involved directly with the NCP.

The Gwich'in still have strong ties to the land and still consume traditional foods; in most cases, traditional food is the main staple of the Gwich'in diet. There has been concern expressed by the Gwich'in about contaminants from long-range and local sources getting into the food chain. The Gwich'in Tribal Council (GTC) recognized that NCP-funded studies dealing with contaminants in the food chain were being done, but there was a need for further studies to be completed in order to address the concerns of the Gwich'in. There was also the need to communicate contaminants information to the Gwich'in, and information about Gwich'in concerns to the NCP. The GTC developed an RCC position, staffed by Norman Snowshoe, in order to address the concerns of the Gwich'in through participation in the NCP.

Activities in 1999-2000

The activities carried out by the RCC in the first year, 1999-2000, were focussed on introducing the NCP to the Gwich'in. Community workshops and public information sessions, with resource people affiliated with the NCP, were held in the Gwich'in communities. Information packages were sent out explaining the role that the NCP has in the studies being conducted. The packages also included information on past and current studies. The RCC attended local meetings in each community to hear and address concerns and to pass on information related to contaminants. Furthermore, the RCC was involved in all activities addressing local source contaminant issues in the GSA, including the collection and testing of fish species and the remediation of an abandoned Shell Oil exploration site (Table 1).

Results

The RCC has raised the awareness of the Gwich'in to the activities of the NCP by holding workshops, collecting and presenting information, and attending local Renewable Resource Council Meetings.

The RCC acquired information related to the consumption of traditional foods near Fort McPherson by collecting and testing fish for contaminants. This has given the people of Fort McPherson assurance that the fish are safe to eat. This analysis was brought on by concerns from people of Fort McPherson about possible contamination from an abandoned Shell Oil exploration site.

The establishment of the RCC position has provided the Gwich'in with coordination capacity to address Local Contaminant Issues. The RCC has also participated in and provided information to the Gwich'in on other federal environmental programs including Climate Change, Cumulative Effects Assessments, Cumulative Impact Monitoring and the Yukon Development Assessment.

Discussion and Conclusions

Within the past year, the Gwich'in RCC has been active in introducing the NCP and information on contaminants to the Gwich'in. This information is very important to the Gwich'in because of the their continued reliance on traditional foods, and the Gwich'in foresee many opportunities for future projects funded through the NCP. The role of the RCC is significant to the Gwich'in as it is a position that is dedicated solely to environmental issues. Finally, because of the continued use of traditional food, the Gwich'in will continue to need information and reassurance regarding the consumption of their traditional foods.

In 2000-2001, the focus of the RCC will be on the development of programs to collect baseline data from the GSA and consultation with the Gwich'in Renewable Resource Councils. The results of these consultations will be the development of proposals to address local source contaminant concerns; these will be submitted to the NCP.

Expected Project Completion Date

The RCC position in the GSA has been renewed for 2000-2001 and thus the work of the RCC is ongoing.

Table 1. Regional Contaminants Coordinator Activities, March 31, 1999 - April 1, 2000

Activity	Location	Date
Community Workshops/Introductions to NCP		
Fort McPherson Renewable Resource Council Meeting – Introduction to NCP	Fort McPherson, NT	August 3, 1999
Meeting with Mayor/Chief Charlie Furlong – Introduction to NCP	Aklavik, NT	August 30, 1999
Aklavik Hunters and Trappers Committee Annual General Meeting: NCP Update	Shingle Point, Yukon	July 23-26, 1999
Renewable Resource Council Meeting – Introduction to NCP	Inuvik, NT	September 13, 1999
Aklavik Renewable Resource Council Meeting – Introduction to NCP	Aklavik, NT	September 15, 1999
Renewable Resource Council Meeting – NCP Update	Tsiigehtchic, NT	November 30, 1999
Contaminants Workshops in the Mackenzie Delta	Inuvik, McPherson, Tsiigehtchic, NT	January 4-7, 2000
Addressing Local Contaminants Concerns		
Peel-Caribou River Contaminated Site Working Group Meeting	Whitehorse, YT	August 10, 1999
Peel -Caribou River Contaminated Site General Meeting with Dr. Dennis Trotter AGRA	Fort McPherson, NT	August 25, 1999
Fish Processing for Contaminant Testing Training	Whitehorse, YT	September 3, 1999
Beaufort Delta Regional Trappers Workshop	Inuvik, NT	September 8-10, 1999
Fort McPherson Bioaccumulation Interviews with Christine Wright	Fort McPherson, NT	August 13, 1999
Tsiigehtchic Bioaccumulation Interviews with Christine Wright	Tsiigehtchic, NT	August 17, 1999
Fort McPherson Renewable Resource Council Meeting-Peel Caribou River Contaminated Site Update	Fort McPherson, NT	September 14, 1999
Peel-Caribou River Contaminated Site Assessment	Peel-Caribou River, YT	September 18-20, 1999
Meeting with Sam Stephenson, DFO-NCP Fish Proposal	Inuvik, NT	October 13, 1999
Peel-Caribou River Contaminated Site Working Group Meeting	Whitehorse, YT	December 2, 1999
Training		
Effective Strategies Training Course	Winnipeg, Manitoba	September 22-24, 1999
RCC Training Course	White Rock, BC	September 27-28, 1999
Cumulative Effects Assessment Training Workshop	Yellowknife, NT	November 15-17, 1999
GIS Course ARCVIEW	Inuvik, NT	January 14-16, 2000
Other NCP Activities		
Northern Contaminants Program Results Workshop	White Rock, BC	September 28-October 1, 1999
NWT Environmental Contaminants Committee Meeting	Yellowknife, NT	February 3-4, 2000
Meeting and Addressing Other Environmental Issues		
Gwich'in Land and Water Board Public Hearing Summary	Fort McPherson, NT	July 12, 1999
Wildlife Act Changes – RWED	Inuvik, NT	October 26 and November 8, 1999, March 17, 2000
Mackenzie Valley Cumulative Impact Monitoring Program Working Group Meeting #6	Yellowknife, NT	November 18, 1999
Gwich'in Land and Water Board Technical Advisory Committee Meeting	Inuvik, NT	November 30, 1999
Cumulative Effects Assessment and Management Workshop	Yellowknife, NT	December 6-10, 1999
Regional Renewable Resource Council Meeting	Aklavik, NT	January 20, 2000
Arctic Borderlands Ecological Monitoring Meeting	Old Crow, YT	February 21-24, 2000
Climate Change Impacts and Adaptations Workshop	Yellowknife, NT	February 27-29, 2000
Cumulative Impact Monitoring Program Working Group Meeting	Yellowknife, NT	February 01-02, 2000
National Roundtable on the Environment and Economy (NRTEE)	Yellowknife, NT	March 29-30, 2000

Contaminants Found Me: a Science Curriculum for Yukoners

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Abstract

This was the final year of a two-year curriculum development project that connects students, grades 3 to 10, to concepts of contaminants, long-range atmospheric transport, biomagnification, bioaccumulation and food choice. This project developed multi-media classroom tools (posters, a music CD, a video, curriculum resource books) that assist teachers and promote student understanding of contaminants. Materials were developed specifically to match the Yukon Department of Education learning outcomes mandated in science Integrated Resource Packages. These materials are available from the Department of Indian Affairs and Northern Development regional office in Whitehorse, Yukon (Phone: 867-667-3272) and at the Yukon Department of Education Learning Resource Centre, also in Whitehorse.

Objectives

1. To build upon the *Contaminants in Northern Canada* science curriculum developed by the Métis Nation-Northwest Territories Contaminants Education Program by creating a science curriculum package with Yukon perspectives.
2. To develop curriculum topics specific to Yukon.
3. To match the learning materials to the Yukon Department of Education's science curriculum.
4. To involve Yukon teachers and students in developing curriculum activities for Yukon classrooms.
5. To focus concepts in the context of Yukon First Nations and local people, wherever possible.

Introduction

Following the 1994 Northern Contaminants Program (NCP) results workshop, the NCP mandated that education and communications activities were to be important components of its program. In 1995 and 1996, the Métis Nation-Northwest Territories Northern Contaminants Education Program produced a three-volume science education curriculum package addressing the topics Environmental Concerns, Nuclear Energy, Northern Food

Chains and Webs, and Pulp and Paper (Métis Nation-Northwest Territories 1995a, Métis Nation-Northwest Territories 1995b, Métis Nation-Northwest Territories 1996). Concepts explored in that science curriculum package are relevant to all of northern Canada but are illustrated within frameworks that are particular to the Northwest Territories and Nunavut. Yukon teachers, however, have a preference to illustrate concepts in frameworks specific to the Yukon and, wherever possible, to present information with relevance to Yukon First Nations.

Material presented in the Métis Nation-Northwest Territories Northern Contaminants Education Program curriculum match the junior curriculum strands mandated in the Northwest Territories. Though it may seem easy to lift materials from one curriculum to another, this is usually not the situation. Schools are structured differently among regions and learning objectives vary. This can affect the presentation, distribution and use of material. Because of this, new materials had to be developed to fit the curriculum in the Yukon.

Materials were developed in the Yukon in collaboration with Yukon teachers and students, with support from the Yukon Department of Education and Yukon Teachers' Association. The curriculum development process, in itself, became an education process as students and teachers learned about concepts highlighted in the curriculum while experiencing the curriculum and curriculum development. Curriculum development and implementation processes will continue after the completion of this project.

In 1997-1998, background research was conducted, and possible topics to be used as frameworks for learning concepts were selected. The following agencies were contacted for cooperation and support for curriculum development in the classroom: Yukon Department of Education; Yukon Teachers' Association; Yukon Native Teacher Education Program (Yukon College); Yukon Environmental Educator's Association; and administrators or teachers in all Whitehorse area schools.

In 1998-1999, ideas were taken into a number of classrooms. Visits to the classroom took the shape of single presentations, field trips and camp-outs, or successive classroom visits. Some classroom visits were conducted with the cooperation of scientists or specialists conducting research on specific topics related to contaminants. The majority of these visits happened during the spring and fall of 1998.

Curriculum materials began to take form in late 1998. Ideas for a series of three posters using "Monty the Moose" to introduce curriculum topics were developed with illustrators (Aasman Design) and approximately 200 Yukon students. Illustrations to be used as graphics for the written text were developed by Tanya Handley (illustrator) and tested as overhead transparencies in numerous classrooms. Ideas for the video *Contaminants Found Me* were developed into a storyboard and script with N. de Graff and students of the grade 9/10 M.A.D. (Music, Art & Drama) program. The music for the video was cut to compact disc under the direction of musician Andrea McColeman and the grade 9/10 M.A.D. program. A demo video of *Contaminants Found Me* was prepared by A.V. Action Yukon and a second video titled *Acid Rock! (drainage)*, was drawn from rough footage by Treeline Productions. All artistic productions have been developed with students and tested in the classroom as tools for the presentation of scientific concepts.

Late in the fall of 1998, the curriculum developers commenced classroom visits to community schools. They also began working with elders, Native language instructors, and the Department of Education First Nations consultants in an effort to connect the curriculum to First Nations world views.

Activities in 1999-2000

This was the final year of the project. Classroom visits were extended by request. Visits to rural community schools were quite extensive and classroom visits to Whitehorse schools continued to the near end of the 1999-2000 school year.

The *Contaminants Found Me: A Science Curriculum for All Yukoners* was prepared for publication. Curriculum resources *Book One: Grades* 3 – 6, *Book Two: Grade* 7, *and Book Three, Grades* 8 , 9 *and* 10 and science kits were made ready for distribution. *Monty the Moose*, (a three poster series), and *Contaminants Found Me* (a music compact disc) were prepared for distribution. These materials were available at the Yukon Teachers' Convention in September 1999, the NCP Results Workshop in September 1999 and the Circumpolar Women's Conference in November 1999. The supply of all materials produced was exhausted early in November 1999. A second printing was made available for distribution before the end of 1999.

Video production was another component of the project this year. The video *Contaminants Found Me* was reviewed by students and teachers in over twenty classrooms before the final edit and Yukon Contaminants Committee approval.

A second video *Acid Rock! (drainage)* was prepared by two Whitehorse students and Treeline Productions. This video is a student discussion of Acid Mine Drainage and some properties of thiobacillus ferrooxidans. The video was reviewed by researchers knowledgeable in this particular field and by students and teachers in eight classrooms. The video was presented in conjunction with the science kits and *Book Two: Grade* 7.

Results

Almost every school in the Yukon expressed an interest in this project. Most Yukon schools now have a curriculum package, which includes posters, a music CD, a video, curriculum resource books. Requests for materials and classroom have been greater than anticipated or projected.

Discussion

This project commenced in October 1997 and was scheduled to conclude in March 2000. Additional requests for materials and class visits extended this project to May 2000.

This curriculum development project has been an interactive process involving many Yukon teachers, students, First Nations educators, scientists and specialists. Concepts developed in the curriculum package are a result of numerous classroom trials. These trials provided direction in determining the best possible approach in educating Yukon students and teachers about contaminants. As classroom sessions evolved, a sense of teacher professional development took form. Teachers learned about contaminants in the Yukon as well as gained a perspective on connecting society, environment and science in the classroom.

The videos are an exciting dimension to the project exemplifying student world views. Students are socially connected to others of similar ages. Students receiving knowledge from students is an empowering process that gives students a strong sense of connecting scientific concepts to learning.

Although the materials have been introduced in many Yukon classrooms, the curriculum development and implementation process of this project is only a beginning. Ideally, materials should be followed and revised on an ongoing basis. A second publication of all these materials should be a future consideration, as well as future classroom visits. A quasi teacher professional development day bringing together researchers, teachers and a select group of students could be an ideal time to address future modifications.

Materials are also available at the Yukon Department of Education teacher resource library. A professional development day whereby teachers can interact with researchers would be an appropriate next step.

Expected Project Completion Date

May 31, 2000

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Yukon First Nations: Draft Cancer Communications Package

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Abstract

Questions have been frequently asked at Northern Contaminants Program meetings and workshops held in the Yukon about the apparent increase in incidences of cancer among Yukon First Nations and whether this increase was related to environmental contaminants. In response to these questions, and at the recommendation of Community Health Representatives and other health workers, a plain language information package about cancer was produced. This package includes 17 frequently asked questions, and provides answers to them. These include general questions, such as "What is cancer and how does it develop?" as well as questions specific to the Yukon population, such as "Who is getting cancer in the Yukon?" "What kinds of cancers are they getting?" and "What are major risk factors for cancer in the Yukon?" The next phase of this work will be to communicate this information to communities. Partnerships are currently being explored with cancer-related organizations and others to complete this phase.

Objectives

Long-term

- To achieve a healthier Yukon First Nations population.

Short-term

- To develop a draft communications package about cancer for Yukon First Nations.

Introduction

First Nations in the Yukon have been asking questions for a long time about how disturbances to the environment may be affecting their health. Since early in the Northern Contaminants Program (NCP), workshops have been held in order to share information about environmental contaminants. At these workshops, people have been asking questions about contaminants and their health effects, particularly about cancer.

The most frequently asked question is *"Why is cancer on the rise in Yukon First Nations communities, and is it related to environmental contaminants in our traditional foods?"* In an attempt to answer these questions, Community Health Representatives (CHRs) and other frontline health workers have strongly recommended to NCP managers in the Yukon that available information, particularly the Yukon Cancer Registry, be reviewed and summarized in plain language.

This has resulted in the need to synthesize available cancer data that is collected in the Yukon and from elsewhere, and to develop a communications package to begin to answer some of the Yukon First Nations questions.

Activities in 1999-2000

In order to produce an information package about cancer that would be useful to Yukon First Nations, we gathered relevant resources and organized a project advisory group. This advisory group was put together to guide the development of the package, to review draft materials and to ensure that government and First Nations health officials were confident from their perspectives that the information was technically accurate as well as readily understandable, and would not generate undue concern.

Work has been done in the Northwest Territories (NWT) to produce a large and comprehensive cancer information package (Dene Nation 1999). This package was reviewed for its relevance in addressing information needs in the Yukon. Permission was given by the Dene Nation for excerpts from this package to be included in the Yukon package. It was agreed by the advisory group that the approach needed in the Yukon at this time was different than that taken in the NWT, with the focus being on brief, plain language answers to frequently asked questions. In the future, there may be a need for detailed information, as contained in the NWT package, so the Yukon and NWT will continue to share communications in this regard.

The Yukon Cancer Registry was reviewed in collaboration with health officials in order to evaluate the types of information that it contains relative to the questions that have been asked at past workshops and meetings. These questions include: What kinds of cancers are occurring among people in the Yukon? Who is getting cancer? How are cancer rates changing over time? and What is known about various risk factors?

We interviewed an elder, Mrs. Ida Calmegane, in her home in Tagish in the spring 2000 to hear about changes that she has seen over the years with people dying of cancer. At the beginning of the interview, Mrs. Calmegane showed us memorials of people who had died of cancer in the last seven to eight years. There were pictures of young people as well as elders who had died from various types of cancer. It was a poignant reminder of how this disease has affected so many people in the small First Nations communities in the Yukon. It also helped to understand the urgency of the concerns of those who have been asking questions about cancer at different contaminants workshops and meetings over the years.

The following questions were agreed upon to be the focus of this communications package:

- What is cancer? How does it develop?
- Who is getting cancer in the Yukon? What kinds of cancers are Yukoners getting? Why is cancer increasing among Yukon First Nations?
- What are the major risk factors for cancer in the Yukon? How can people lower their risks of getting cancer? Is cancer hereditary? What can I do to prevent cancer?
- What is the cancer mortality rate for Yukon First Nations, and has that changed in the past 10 years? How many people died in the past 10 years, and from what? How many people have been living with cancer in the Yukon over the past 5 years?
- What is the life expectancy of Yukon First Nations relative to other Yukoners? to other Canadians?
- How has the age structure of Yukon First Nations changed over the past 40 to 50 years? What does this mean for present and future cancer rates in the Yukon?
- What information resources are available to help people understand about cancer and the situation in Yukon communities? Who can people contact for further information about cancer?

Answers to these questions were drafted, reviewed and revised by the advisory group. Digital images were produced to characterize risk factors and other images were collected to illustrate healthy lifestyle choices. Attempts were made to transmit this information in a holistic way, respecting the complete and integrated First Nations perspective, and using plain language.

Results

The results of this work have been compiled into a booklet and images that are intended to answer frequently asked questions about cancer by Yukon First Nations, and to increase peoples' awareness about how healthy lifestyle choices can reduce the risk of cancer. This information now needs to be shared with communities.

Discussion and Conclusions

This work has been the first step in producing a plain language cancer information package for Yukon First Nations. This meets a need identified by community representatives at various contaminants meetings, as well as by CHRs and other health workers.

The exchange of this information with communities in the Yukon, as well as identifying follow-up that may be required to address additional questions, are the next steps. Also, identifying connections already in communities will be important for the development or expansion of local community empowerment with respect to the subject of cancer, particularly if youth and elders can work together. We are exploring partnerships with cancer-related organizations for funds to travel to communities, as well as identifying additional expertise to facilitate the ongoing exchange of information.

Staff from the Whitehorse office of the Canadian Cancer Society are traveling to each community in the Yukon in the fall to talk about cancer with community members. This would be an ideal time to present and discuss the information in the booklet. We are currently seeking funds to undertake this travel.

Options for the piloting and communication of the draft package have been identified. These include holding community workshops, producing a series of pamphlets, talking about the frequently asked questions on the radio, posting one question per week on the rolling ads on TV, specific training for CHRs and other health workers, producing posters and involving the Health Commissioners in each community. These are all good ideas and funding is currently being pursued to implement some or all of them.

Expected Project Completion Date

The project, as defined by the proposal which was funded, is now complete, although there is still work to be done to ensure that the communities' concerns and questions that initially generated this project are addressed to their satisfaction.

The extent to which the NCP contributes to future developments or proposals in this regard will be largely determined by program priorities, and other sources of funding will continue to be actively sought to meet communities' information needs about cancer.

Acknowledgments

We would like to thank the members of the advisory group for their significant contributions to this project, and especially to Mrs. Ida Calmegane for sharing her wisdom and ideas with us.

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Eastern Arctic Contaminants Tour

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Abstract

> *I'm really glad that you guys are here today because before this meeting, I've been concerned about eating country foods because of the contaminants. This meeting put a smile back on my face. And I can continue eating country food again.*
>
> – Elder, Hunters and Trappers Association, Northern Contaminants Program workshop in Grise Fjord

Two small teams, each consisting of a program representative, an Aboriginal partner, a health specialist and a scientist, visited six communities between mid-November and early December 1999 to conduct community meetings on contaminants. Topics of discussion at each meeting included: where contaminants come from, contaminants in the ecosystem and food chain, contaminants and human health, and ongoing activities. The tour met its objectives and also helped to remind team members of the importance and relevance of the work the NCP is doing.

Objectives

1. To communicate the goals, objectives and priorities of the Northern Contaminants Program (NCP) Phase II to seven Inuit communities in Nunavut.
2. To outline recent and current contaminant projects in Nunavut as well as to relay the findings of the NCP Phase I.
3. To identify issues and concerns of communities in Nunavut with regard to contaminants in the environment for the development of a Nunavut monitoring plan.

4. To assist individuals and communities in Nunavut in making informed decisions about their use of country foods.

Introduction

The most common type of workshop used to convey Northern Contaminants Program (NCP) research results and program information has involved bringing selected community representatives to a central location, providing them with information regarding contaminants and the NCP and then sending them home. It had been the hope that those representatives would spread the information to the rest of the communities. This was realistically an unfair burden to place on the community representatives because most were not in a position to talk to every interested person nor was it usually their job to do so. Workshop participants would often be overwhelmed with information, which created the potential for the erroneous interpretation of results.

An alternative method of delivering the contaminant and NCP-related information is to send a small, knowledgeable team to a community for a couple of days. Ideally, such a team would consist of a program representative, an Aboriginal partner, a health specialist and a scientist. While the primary means of communication would be a public meeting, the team would take advantage of the time in the community to meet also with the Hunters and Trappers Association, the hamlet council, the schools and any interested groups.

The advantages of sending a team to a community rather than bringing a single representative to an outside workshop are numerous. Not only could more individuals be reached, they would be hearing the message from the source rather than second-hand, and the team would be able to answer most questions immediately. By being in the community and doing more than just the public meeting, it also gives individuals the chance to hear and comment about contaminants more than once. Also, there is a level of comfort afforded the individuals since they are with their families and neighbours which should help make people more comfortable to express opinions or concerns about the program.

Another benefit is the relationship-building that goes on while in the community. By going to the community, people get to see some of the individuals doing the work and this can help to build a better working relationship.

A lesser benefit, but one that should be considered, is the economic benefit to the community. While in the community, the team inputs money into the local economy through accommodations and meals, logistical support (e.g. meeting organization, translation services, hall rental) and souvenir shopping.

Activities in 1999-2000

The primary activity in 1999-2000 was the organization and delivery of seven community workshops in the eastern Arctic in late November and early December, 1999.

Results

In response to the overwhelming interest from communities, the first seven communities that had responded to an initial offer of a visit by a small NCP team were selected for the tour. The tour was carried out over two trips. The first trip was to include Repulse Bay, Igloolik and Hall Beach; the second included Resolute Bay, Grise Fjord, Pond Inlet and Clyde River (Table 1). Unfortunately a raging blizzard prevented the team from reaching Hall Beach. At each community, the team hosted a workshop in the evening and tried to meet with other groups such as schools and Hunter and Trapper Organizations. While it was recognized that these other meetings were important, they were difficult to arrange, especially with a tight travel schedule.

Table 1. Itinerary for the Eastern Arctic Contaminants Tour, 1999

Community	Date
Trip 1	
Repulse Bay	November 14, 1999
Igloolik	November 17, 1999
Hall Beach	Cancelled due to weather
Trip 2	
Resolute Bay	November 30, 1999
Grise Fjord	December 1, 1999
Pond Inlet	December 2, 1999
Clyde River	December 3, 1999

Along with the individual talks of the various speakers, a four-part panel presentation was developed to graphically show the four themes of the workshop: where do contaminants come from (Figure 1), contaminants in the ecosystem and food chain (Figure 2), contaminants as they relate to human health (Figure 3), and ongoing activities (Figure 4). After the tour, the presentation was

Figure 1. Where do contaminants come from?

Figure 3. Contaminants as they relate to human health

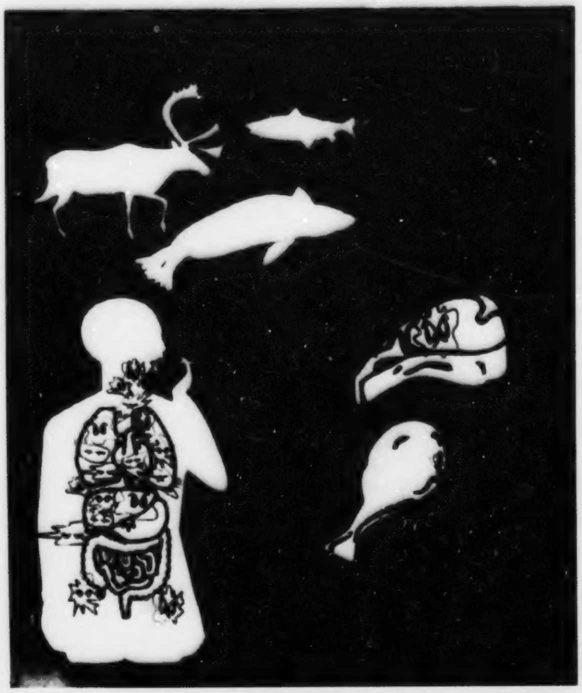


Figure 2. Contaminants in the ecosystem and food chain

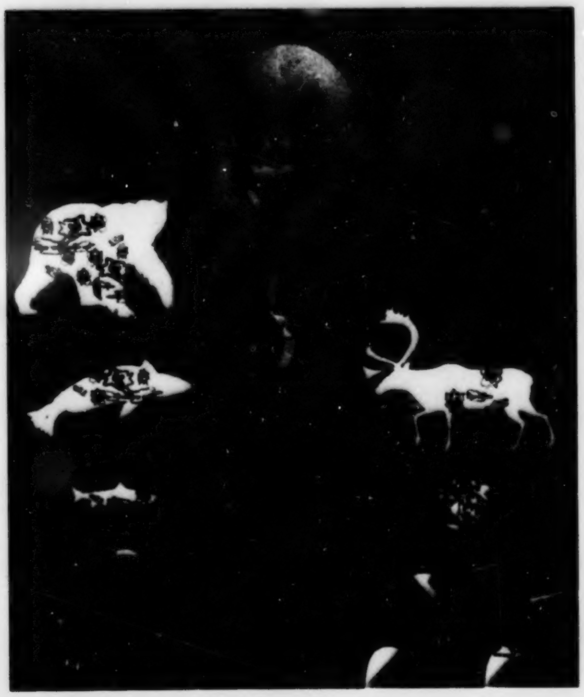
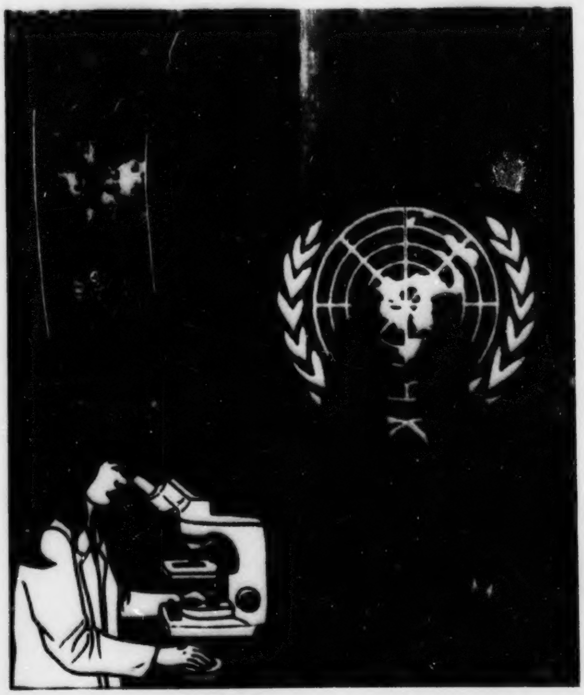


Figure 4. Ongoing activities



reformatted as a poster (Figure 5) and sent to various organizations within each community.

A trip report, which describes each meeting in detail, is available from the project leaders.

Discussion and Conclusions

The tour proved to be a success in every community in which it stopped, with many communities wanting a team to come back at some future date (the sooner the better!). During each workshop, there was a good two-way dialogue which helped everyone understand the issues and concerns. The opportunity to talk to other organizations was not often possible, and is something that should be addressed prior to similar future tours.

As with so many things, practice and experience makes things better, and any future tour will benefit from the experiences learned on this first tour in such areas as advanced communications, workshop design, key messages and displays, as well as souvenir shopping.

One outcome not anticipated from the tour, but perhaps the most valuable outcome for the NCP, was the increased awareness among the team members of the importance and relevance of the work that the NCP is doing.

Expected Project Completion Date

This project is completed and another tour is planned for Fall 2000.

Figure 5. Community contaminants poster

Inuit Response to Environmental Contaminants Information: the Cape Christian-Clyde River Case

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Abstract

The present study focussed on two issues: i) the quality of information received by Clyde River Inuit about the contaminants situation at Cape Christian; and ii) the traditional knowledge-related means and/or indicators used by Clyde River Inuit to evaluate the quality of local resources as food. Following an extensive literature search, field data were collected from Clyde River Inuit during July 1999 and from February-April 2000. Information obtained suggests that the frequency of information flow about the contaminants situation at Cape Christian was sporadic, at best, but has been effective in informing Clyde River Inuit of the potential health hazards present in the immediate area of the former Long Range Navigation station. Clyde River Inuit are less informed about the contaminants risk related to their continued use of marine resources, notably ringed seal, from the waters adjacent to the former station. All Inuit interviewed during both field phases expressed considerable knowledge and concern about the condition of Patricia Bay, especially with regard to the use of its eastern portion near the former village and Ministry of Transportation weather station, and the effect of leaching of various chemicals on the quality of both the local Arctic char fishery and clamming beds. While it is difficult to judge how widely held is traditional knowledge information related to evaluating local food quality, it is clear that local concern about contaminants is not limited to only the two most immediate sites focused upon in this research. A number of views are held on the causes and loci of perceived food problems, suggesting that improved information flow from external sources to communities is important not only to inform residents about specific problems, but also to dispel or correct erroneous "hypotheses."

Objectives

1. To examine various archived records on the Cape Christian clean-up and to conduct interviews with Clyde River hunters regarding their use of the Cape Christian area in order to determine whether information provided to the people of Clyde River about chemical

pollution at the former United States Coast Guard station at Cape Christian has affected the way Clyde River hunters use that area and the adjoining waters and sea ice.

2. To investigate how Clyde River Inuit determine the suitability of harvested animals as food, drawing upon their traditional knowledge regarding the health of individual animals, the signs and/or symptoms that identify an animal as being unacceptable as human food, and Inuit views on the etiological or other causes for the "unhealthy" condition of so identified animals.

Introduction

In 1992, Environment Canada announced that the area adjacent to the decommissioned United States Coast Guard (USCG) Long Range Navigation (LORAN) station at Cape Christian, Baffin Island, was heavily contaminated by polychlorinated biphenyls (PCBs). There was also concern that the site located approximately 20 km from the Inuit community of Clyde River might be leaching PCB-contaminated run-off and groundwater into adjacent Baffin Bay. Over the following three summers, a clean up of the Cape Christian site was undertaken by Environment Canada. This marine area, known locally as Pikaluit, was an important winter sealing and polar bear hunting area for the Clyde River Inuit. Inuit were advised to avoid the lands adjacent to the station area; however, no warnings were issued regarding the consumption of local wildlife. Clyde River hunters have expressed concern about the condition of lands at Cape Christian and the potential hazard of consuming seals and polar bears from within the Pikaluit area. By 1995, a number of Clyde hunters began to express concern both about the condition of the lands at Cape Christian and about the potential hazard of consuming seals and polar bears captured from within the Pikaluit area.

Prior to this project, a pilot study on the presence of PCBs in the foods and diet of Inuit at Broughton Island, East Baffin Island was conducted by Kuhnlein and Kinloch (1988). The results from this study showed that substantial quantities of PCBs were entering the human food system through the consumption of marine mammals. These results frightened the community; local concern was such that for several years the Broughton hunters shifted their seal hunting to areas more distant from the perceived area of contamination and reproductively active women were advised to limit their consumption of certain marine mammal products. Other studies conducted since then have continued to show that the contamination of traditional Inuit foods remains a concern and that local reaction to such information (Usher et al. 1995) can have a disruptive impact on local diets.

This project looks solely at the issue of PCB contamination at Cape Christian and of the Pikaluit area and how this contamination fear has affected certain aspects of Clyde River Inuit subsistence relations.

Activities in 1999-2000

The project was implemented in three phases. Phase I took place during May-June 1999, and involved: i) the collection of federal and Northwest Territories government documents relevant to the Clyde-Cape Christian case; and ii) telephone interviews with members of federal, territorial and Inuit organizations cognizant of the Clyde-Cape Christian situation.

Phase II occured in July 1999 and involved securing final permission from the Namautuq (Clyde River) Hunters and Trappers Organization (HTO) for the research. Subsequently, the following activities were undertaken: a review of notes and/or documents held by the HTO; the formulation of the project interview protocol (developed with Mr. Esa Qillaq); solicitation via community radio of possible project participants; the conducting of recall interviews about community-agency communications and contaminants at Cape Christian; and the pre-testing of the formal research protocol. Pre-testing and file review were delayed nine days until a meeting of the full HTO executive committee could be convened.

Following the securing of committee permission, preliminary data collection began. The results of the pre-test exposed a number of limitations in the protocol; these were corrected during September-October, 1999, and transmitted back to Clyde River for second testing.

Phase III, which involved collecting substantive data pertinent to the project's objectives, was begun by E. Qillaq in late January 2000, and extended until April. Translated (Inuktitut to English) copies of the data were forwarded to Montreal for analysis by G. Wenzel in early May. In total, interviews were conducted with 21 Clyde residents, including five women and 16 men. All interviewees were 30 years of age or older and all males interviewed are or were hunters who actively used the marine environment adjacent to Cape Christian.

Results and Discussion

Archival and other external sources

Locating and retrieving archival information proceeded in several steps. Sources related to contaminants in the Eastern Arctic, in general, were initially searched for through the Arctic Institute of North America's research database (Arctic Institute of North America 1999) and then through electronic searches of holdings in specific libraries and offices in Yellowknife, Montreal, Ottawa and Iqaluit. In addition, information about the Cape Christian facility was sought from sources at the USCG Academy and in Washington, D.C. All such searches were undertaken by the principal investigator over a six-week period spanning May and June. Table 1 details the agencies and/or organizations contacted during this period regarding the Cape Christian contaminants situation.

Considerable information was found to be available on the extent and degree of remediation undertaken concerning organic pollutants (principally PCBs) at the former USCG Cape Christian installation (e.g. Indian and Northern Affairs Canada 1996) and Old Clyde River (Indian and Northern Affairs Canada n.d.). These materials made it clear that Canadian federal government concern about the site was first expressed in 1985 and that clean-up activity at Cape Christian extended into the early 1990s. However, none of the archived materials located provided specific or summary information on the extent or content of the communications that may have occurred between external agencies and organizations involved in the assessment, remediation and follow-up stages of the Cape Christian clean-up and the community of Clyde River. Similarly, no documentation, in the form of minutes or transcripts, of agency-community contacts was present in the files of Clyde's HTO or hamlet office.

Examination of several externally archived documents revealed that contaminants remediation was also undertaken by the Government of the Northwest Territories authorities during the 1990s at the former Ministry of Transportation (MTO) weather station (active until 1982-1983), located across Patricia Bay from Clyde River. This information was incorporated into the revised protocol used during the substantive interview phase of the research.

Because of the absence of information in published or gray sources related to clean-up in the Clyde area, a number of individuals with knowledge of the Cape Christian and/or Clyde MTO assessment and clean-ups were contacted by telephone regarding agency communications with the Clyde community. The sum of these contacts (n = 6) revealed that only one person interviewed had actually participated in the information meetings with the community, however, he was unable to recall whether any formal record of the content of these meetings had been kept.

Clyde River information sources

Community records

Following the disappointing outcome of various document searches and telephone interviews undertaken in or with sources in Ottawa, Montreal, Yellowknife and Iqaluit, a search of available records in the hamlet government, HTO and Sustainable Development offices was initiated after obtaining research permission from the HTO. Unfortunately, this also proved to be unsatisfactory, as apparently no written or audio records were kept of contaminants-related meetings in the community (S. Sather personal communication, M. Audlakiak personal communication).

Clyde River Inuit recall

In lieu of any locally archived material about contaminants information communicated through meetings within the community, a series of recall interviews were conducted between July 8 and July 18 with 21 Clyde River Inuit. Interviews focussed on individuals' memories of any information received about the Cape Christian situation and about any concerns that may have been expressed at contaminants information meetings attended by the interviewees in Clyde River.

The responses elicited through these interviews, despite a general lack of clear memory about the content of specific meetings (at least three such meetings were variously, if vaguely, recalled, the most recent of which appears to have taken place circa 1995), brought out two substantive points. The first being that the consensus understanding among those interviewed is that Cape Christian (the physical site, including its buildings and the lands immediately around them) was and remains contaminated by PCBs and should therefore be avoided. Second, that there has been no serious impact on the adjacent marine environment. Following this, when individuals were asked if, for instance, they subjected ringed seals from the waters off Cape Christian to closer scrutiny than seals captured from more distant areas, in terms of either externally discernable anomolies or as to the "wrong" appearance of internal organs or fat, the unequivocal answer was "No."

The summer 1999 and winter 2000 interviews brought out several further points. The first was that, with respect to the overall condition of the Clyde environment, nine of the 21 Inuit interviewed, independent of interviewer prompting, noted as a concern the possible continued transport of contaminants from the former MTO meteorological site at "Old Clyde" (closed 1980) into Patricia Bay and the possible continuing adverse impact of this leakage on Arctic char from the "MOT Lakes" during their sea-run phase. In this regard, the nine respondents hypothesized that "chemicals" from the former MOT operation had caused a precipitous decline in the size of the local run over the last two decades and four hunters further suggested that "...every summer fish are caught with eye problems."

A second unexpected point was that almost all of the 21 interviewees (n = 16) cited contaminants cases from outside the Clyde region (notably the Cape Hooper and Fox Charlie Distant Early Warning [DEW] Line site situations) when asked about their knowledge about the northern contaminants situation. In several cases, this was almost certainly due to the fact that several respondents had close relatives who were employed in remedial operations at these sites. It may also be the case that the northern media have raised general public consciousness about the contaminants situation along the former DEW Line.

Traditional knowledge and contaminants

The paucity of information related to the extent and type of communication received in Clyde about contaminants and the Cape Christian and MOT situations was disappointing, especially as the recall interviews conducted in 1999 provided little amplification about the information exchange that occurred. On the other hand, the responsiveness of Clyde residents interviewed during the July pre-test about the identification of possibly contaminated animals and the sources of these occurrences suggested that the winter 2000 phase of the research concentrate on the second objective of the project.

Following preliminary assessment of the July interviews, a meeting was convened with the executive committee of the Naumatuq HTO. In this meeting, discussion centered on the utility of concentrating the remainder of the research effort on the traditional knowledge aspects of "contaminants," namely what indicators are used by hunters and others in determining the health status of local resources as potential food. After reviewing the archival and recall results and discussing the limitations of both data sets with the executive committee, it was decided that the substantive interview phase should mainly focus on how Clyde hunters and food preparers identify "problem" animals. This traditional knowledge phase of the research, therefore, had the following foci: i) the health status indicators used by hunters when handling animals at the time of their capture; ii) the status indicators used by food preparers during the secondary handling of animals; iii) whether abnormal animals appear to be concentrated in specific areas of the Clyde region; and iv) the "hypotheses" put forward by respondents as to the source causes of unwellness in animals identified as unsuitable.

Species-specific indicators

Results pertaining to the first and second foci above are presented via species categories.

1. Polar Bear ("Nanuk" *Ursus maritimus*)

The most frequently mentioned physical characteristics that the hunters who were interviewed said indicated abnormality in polar bears were very yellow skin, patchy pelage and extreme thinness. With regard to the yellowness of a bear's skin, several respondents made it clear that their reference to coloration was not about fur color and that yellowish fur indicated a bear that is well fed, with the yellowishness being caused by the fur being temporarily stained by seal fat.

Two men and two women also noted that they had, in the course of handling polar bear skins or meat, observed either red spots in the subcutaneous fat (three responses) or white spotting in the meat (one response). All four stated that such spots indicated a "sick" bear that should not be eaten. Further, one male respondent noted that the spotting he had observed was in the animal's fat and thus not related to parasites, which are located in muscle tissue and often give polar bear meat a "sandy" appearance.

2. Ringed Seal ("Natsik" *Phoca hispida*)

The ringed seal is the primary marine mammal food species consumed at Clyde. Two hunters mentioned that they had at various times captured "natsik" with yellowish pelage and that they regarded this as a sign of the animals' unfitness as food. The only other surficial indicator of unwellness in seals mentioned was that any animal with an obvious injury that a hunter might catch would be inedible because the injury's effect on the seal would change its taste, making the meat "sour."

Related to the above, at the time of the July pre-test, an incident occurred at Clyde that stimulated a variety of discussion pertinent to the project. A hunter returned from an area southeast of Clyde with a ringed seal that was essentially "bald," having only a few small and scattered patches of reddish tinted hair on its left thorax area and at the base of its frontal and hind flippers.

This animal evoked considerable discussion about possible causation; opinion as to how this had occurred ranged from "pollution" to harassment by motorized traffic in the area of capture. While no consensus was reached regarding cause, opinion was unanimous that the animal should not be eaten. Further, several of the community's older residents stated that seals in similar condition had occasionally been captured in the past, but no specifics as to when or location were recalled.

Beyond surficial signs of unfitness, both women and men noted that red or yellow spots in the fat of seals was a sign that they should not be eaten. The only opinion about the cause of such fat spotting, however, was that such animals are "sick."

3. Bearded Seal ("Ujjuk" *Erignatus barbatus*)

Bearded seal, while opportunistically hunted for their skins (for the manufacture of sealskin rope), are only infrequently eaten by Clyde River Inuit. However, one respondent was explicit that the same surficial indicators of unwellness as mentioned above for ringed seal apply to "ujjuk."

4. Other Marine Mammals

No information was obtained regarding narwhal, walrus, or harp seal, the other principal marine mammal species harvested by Clyde River Inuit. This may be due to the fact that: i) harp seal is rarely actively pursued and almost never eaten, ii) walrus are taken only very occasionally, most recently, one in 1995; and iii) the harvest of narwhal is highly variable (annual average = 24) and its use for food exclusively focuses on the "muktaaq," which has considerable cultural value at Clyde. It is possible that the premium placed on this product outweighs close scrutiny for physical anomolies, although Clyde River Inuit typically comment on the presence of "sea lice" during narwhal butchering operations.

5. Caribou ("Tuktu" *Rangifer tarandus*)

All the hunters interviewed made it clear that they clearly distinguished natural "problems," like brucellosis, from those caused by "pollution." No one interviewed associated any of the latter type of problem with contaminants originating from either Cape Christian or the Clyde MOT sites. The reason invariably given for this is that caribou are known to have disappeared from the southern forelands, where both sites are located, in the early 1950s and caribou, therefore, are seen as never coming into proximity to these sources.

Six hunters, however, noted that they had captured caribou that were unseasonably thin in terms of both pelage and expected amount of body fat, and that this condition may have been the result of the animals having foraged on contaminated fodder. The one area where captures of animals in such condition seem to have concentrated was on the western side of Baffin Island between Nauja Bay-Baird Peninsula and the Barnes Ice Cap. It was generally speculated that animals feeding near the Longstaff Bluff DEW Line station have been affected by contaminants originating from that site.

Hunters also speculated that caribou suffer from fuel-contaminated fodder when they feed near the many aviation gasoline caches that dot parts of the interior Clyde area. Again, the interviewees seemed to be of the opinion that animals so affected were generally found west and south of the Barnes Ice Cap.

Virtually all information obtained on caribou was drawn from male respondents, with little significant addition from the women. This "bias" has its roots in the fact that caribou are generally harvested at considerable distance from Clyde River and almost invariably undergo "dressing" in the field.

6. Arctic Char ("Iqaluk" *Salvelinus alpinus*)

Nearly every person interviewed made reference to having encountered Arctic char from Patricia that exhibited significant anomolies. In every instance, the cause of these conditions was considered to be directly linked to the leaching of various chemicals spilled during weather balloon launches that took place from the MOT station on the east side of Patricia Bay. As Forbes (1993) indicates, aerial photographs of the former MOT-Inuit village site show that, from 1960 onward, considerable chemical leaching flowing from the spill site toward and into Patricia Bay occurred.

While Clyde River Inuit generally note that Arctic char originating in the MOT Lakes system, and which are taken from Patricia Bay, have declined in size, it is likely that this is the result of the heavy utilization of this fishery over the last 40 years. However, all the respondents interviewed noted that fish captured in Patricia Bay often exhibit bulging eyes, distended livers and generally possess less subcutaneous fat after the sea-run than char taken from other systems in the Clyde region.

Of all the species-specific interviews conducted during the winter phase of interviewing, it was those concerned with Arctic char that elicited both the greatest degree of concern and uniformity of causation. All respondents noted that fish taken from Patricia Bay must be scrutinized for signs on abnormality and that older residents frequently make note of this to younger harvesters.

7. Clams ("Ammuumajuq" species unknown)

The waters of Patricia Bay are also important to Clyde River Inuit for the harvesting of clams. While "ammuumajuq" are a minor component of the Clyde resource inventory, they are gathered from the waters immediately in front of the former MOT site.

Six respondents (two men and four women) made explicit reference to the belief that the productivity of this area and the quality of the resource obtained has undergone gradual deterioration. Noting that clamming is very much a minor activity for Clyde River Inuit, these interviewees stated that there appear to be fewer clams than were present formerly along the beach area fronting the old village and that a significant percentage of the bivalves

Table 1. Cape Christian, Clyde River Project Archival Sites and Contact Log

Organization	Location	Interview conducted with agency representatives
Nunavut Research Institute	Iqaluit, Nunavut	Yes
Indian and Northern Affairs Canada	Iqaluit, Nunavut	No
Qiqiqtaaluk Corporation	Iqaluit, Nunavut	Yes
Department of Sustainable Development, Government of Nunavut	Iqaluit, Nunavut	Yes
Naumataq Hunters and Trappers Organization	Clyde River, Nunavut	No
Wildlife Division Office, Department of Sustainable Development	Clyde River, Nunavut	No
Contaminants Division, Indian and Northern Affairs Canada	Yellowknife, Northwest Territories	Yes
Environmental Protection Office, Department of Resources, Wildlife and Economic Development, Government of the Northwest Territories	Yellowknife, Northwest Territories	Yes
Prince of Wales Heritage Centre Documents Centre	Yellowknife	
LDS Engineering Ltée	Montréal, Quebec	Yes
Documentation Centre, Canadian Museum of Civilization	Hull, Quebec	No
Environmental Sciences Group, Royal Military College	Kingston, Ontario	No
Library, United States Coast Guard Academy	New London, Connecticut, U.S.A.	No
United States Coast Guard Liason Office	Washington, D.C., U.S.A.	Yes

that are gathered are dead. Each of the six saw a direct link between the eroded quality of this resource and the effluent that is believed to now be leaching into Patricia Bay from the "old site."

Site issues

In both the preliminary and winter interview phases, three areas were identified as sites of concern. The interviews made it clear that Clyde residents share considerable concern over the possible persistent environmental effects of contaminants at both Cape Christian and the former MOT.

With regard to Cape Christian, while it is clear that there is widespread awareness of the remediation efforts that have taken place, there remains strong concern about the condition of the remaining buildings and the surrounding lands, with frequent note made of the need to avoid the immediate LORAN site or areas where refuse was buried at the time of decommissioning. Less concern was evinced, however, with respect to the marine environment and resources other than polar bear, which are known to spend time near the buildings. The only avoidance noted and practiced, with regard to marine resources from near Cape Christian, is that hunters restrict their shoreline hunting by not venturing closer to the known area of contamination than the southeastern end of the Cape Christian airstrip.

More concern was apparent among respondents as to the possible continuing effects of contaminants released, and leaching into Patricia Bay, from the decommissioned MOT weather station. Interviewees emphasized that Clyde residents generally avoid fishing on the eastern side of the bay and that the two main food items harvested from the bay, Arctic char and clams, require close examination before actual use.

The only other site mentioned, with any frequency, as possibly affecting local resources was Longstaff Bluff, with regard to caribou. Here, knowledge was more restricted as relatively few of the interviewees (n = 3) had direct experience with caribou from that area.

It should also be noted that a few respondents made mention of more distant situations, notably the Cape Hooper and Fox Charlie DEW line sites. However, only one person cited direct knowledge of either place and only because she had worked with a recent remediation team. Several residents also mentioned the possible deleterious effects of the run-off originating from the "new" Clyde dump, which empties through several small streams into the western side of Patricia Bay, some several hundred metres south of the community (Forbes 1993). In this regard, the most significant indicators of "pollution" noted were lusher vegetation outlining the dump's discharge plume and the profusion of algal growth on the rocks located near the area of discharge.

Conclusions

The present study focussed on two objectives: 1) the quality of information received by Clyde River Inuit from external sources about the contaminants situation at Cape Christian; and 2) the traditional knowledge-related means and/or indicators used by Clyde River Inuit to evaluate the quality of local resources as food.

The information obtained from archival and Clyde River sources about the first research issue was disappointing, suggesting that, at best, the frequency of information flow about the contaminants situation at Cape Christian was sporadic. Nonetheless, it would appear that this has been effective in informing Clyde River Inuit about the potential hazards to human health present in the immediate area of the former LORAN station. The Inuit were clearly cognizant of the hazard represented by the de-commissioned buildings and the terrestrial environment in their immediate vicinity.

On the other hand, the data suggested that Clyde River Inuit are less informed about the contaminants risk related to their continued use of marine resources, notably ringed seal, from the waters adjacent to the former station. While interviewees noted that they did not conduct spring shoreline hunting from the perceived "hot" area of contamination, they do continue to seal hunt along the shore area immediately below the station, despite the fact that the prevailing current is from the station to this hunting area.

In contrast, all Inuit interviewed during both field phases of the project expressed considerable knowledge and concern for the condition of Patricia Bay, especially with regard to the use of its eastern portion near the former village and MOT weather station. Particular reference was forthcoming to the effect that the leaching of various chemicals once employed in MOT operations have had on the quality of both the local Arctic char fishery and clamming beds.

With respect to the second research issue, that of traditional knowledge regarding food quality, considerable information was gained, with respect to the kinds of physical indicators used by Clyde River Inuit, to evaluate the quality of a variety of terrestrial and marine food species. While it is difficult to judge from the limited sample employed how widely this traditional knowledge information is held, the data make it clear that local concern about contaminants is not limited to only the two most immediate sites focused upon in this research.

The research on traditional knowledge information about food quality also elicited a number of views on the causes and loci of perceived food problems. While these views were highly speculative, they do suggest that improved information flow from external sources to communities is important not only to inform residents about specific problems, but may also serve to dispel or correct erroneous "hypotheses."

Project Completion Date

March 31, 2000

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International Policy

Canadian Arctic Indigenous Peoples Against Persistent Organic Pollutants: Support for International Action

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Abstract

The five northern Aboriginal partner organizations of the Northern Contaminants Program (Council of Yukon First Nations, Dene Nation, Métis Nation-Northwest Territories, Inuit Circumpolar Conference, Inuit Tapirisat of Canada), jointly participate as a coalition, the Canadian Arctic Indigenous Peoples Against Persistent Organic Pollutants, in international negotiations sponsored by the United Nations Environment Programme to finalize an international convention to eliminate key persistent organic pollutants (POPs). A representative of these organizations sits on the Canadian delegation to the negotiations, and representatives of each organization attend negotiations as observers, taking advantage of the Inuit Circumpolar Conference's status as an official observer to the United Nations Economic and Social Council. In the 1999-2000 fiscal year, global POPs negotiations took place in Geneva and Bonn. The coalition participated in both meetings.

Objectives

1. To raise international awareness of the concerns of Canadian Arctic Aboriginal peoples about contaminants.
2. To ensure Canadian Arctic Aboriginal people participate fully in domestic and international contaminant decision making.
3. To ensure the eventual international agreement on persistent organic pollutants protects Canadian Arctic Aboriginal people.

Introduction

The adverse effects of contaminants on both environmental and human health is of great concern in the North and as such, the North has a great interest in the success and effectiveness of the current international initiatives to reduce the presence of contaminants in the northern environment. For these reasons, it is crucial that northern Aboriginal groups understand the issues, participate in the development of the Canadian negotiating positions and continue their role as an integral component of the Canadian and international effort.

In the spring of 1997, with support from Environment Canada and the Department of Indian Affairs and Northern Development-led Northern Contaminants Program (NCP), the five northern Aboriginal organizations (Council of Yukon First Nations [CYFN], Dene Nation, Métis Nation-Northwest Territories, Inuit Circumpolar Conference [ICC] Canada, Inuit Tapirisat of Canada [ITC]) joined to form the Canadian Northern Aboriginal Peoples Co-ordinating Committee on Persistent Organic Pollutants, later to be re-named the Canadian Arctic Indigenous Peoples Against Persistent Organic Pollutants (CAIPAP). Through this coalition and a part-time technical advisor, the members of CAIPAP raised the level of awareness in the North concerning international initiatives underway on persistent organic pollutants (POPs) and brought the concerns of Arctic residents on the contaminants issue to the global community. CAIPAP has expanded to include representation from the Nordic Saami Council, the Russian Association of Indigenous Peoples of the North (RAIPON) and the Aleut International Association.

CAIPAP participates in meetings with the Canadian delegation to recommend positions and priorities that reflect Aboriginal and northern interests. As well, this coalition cooperates with the Indigenous Environmental Network, the International POPs Elimination Network, World Wide Fund for Nature, the Canadian Arctic Resources Committee and other organizations. The coalition is also cooperating with the North American Council for Environmental Co-operation to promote the control of POPs that end up in the Arctic.

All Canadian members of CAIPAP use their partnership role in the NCP as a base for involvement in international aspects of POPs management and elimination. In support of the coalition's international activities, ICC Canada and ITC have also concentrated attention on the national and international media, and participated in amendment of domestic legislation and policy including the *Pest Management Control Act* and the *Canadian Environmental Protection Act*.

Through the efforts of the CAIPAP, the concerns of Arctic residents about contaminants are being heard by the global community and northern Aboriginal people are influencing the international process to eventually eliminate POPs from the northern environment.

Activities in 1999-2000

During the last year, the five Canadian members of CAIPAP – CYFN, Dene Nation, Métis Nation-Northwest Territories, ICC Canada and ITC – participated in international negotiations in Geneva (September 1999) and Bonn (March 2000) toward a global convention to eliminate and/or manage key POPs of concern to Northerners.

Discussion and Conclusions

The coalition, CAIPAP, has proven itself to be an effective mechanism not only to involve Canadian Indigenous people in international negotiations on POPs, but also to assist Indigenous people in Russia and Scandinavia to better understand and participate as well. Representatives of RAIPON joined the coalition in both international negotiating sessions in the 1999-2000 fiscal year.

At a political level, Sheila Watt-Cloutier, President of ICC Canada, is the acknowledged spokesperson for CAIPAP, as is Shirley Adamson, ex Grand Chief of CYFN. Political statements and positions taken by the coalition are approved by all coalition members, as is the basic platform and position developed by staff. Staff from all five members are supported by Stephanie Meakin, an Ottawa-based consultant. The ability of the coalition to participate in international negotiations is very much an outgrowth of the excellent relationship between the members established through shared management of the NCP and by joint participation on the board of the Centre for Indigenous Peoples' Nutrition and Environment at McGill University.

The coalition has developed a broad position on substantive issues being addressed in the international negotiations. In particular, the coalition insisted upon a convention that is comprehensive, verifiable, and rigorously implemented and that will lead to elimination rather than perpetual management of offending POPs. Translating these principles into operational language in the forthcoming convention is a challenging, ongoing task. To do this requires the coalition to engage in technical arguments during negotiations and in intersessional processes. CAIPAP does so on a number of fronts, including, for example, timetables for assumption of phase-out obligations, funding implementation of the convention, trade restrictions, capacity building in developing nations, and effectiveness of the compliance regime. Our position papers deal with these and other issues in detail. Major correspondence by ICC Canada and CAIPAP on this issue may be found on the ICC Canada Web site of at www.inuitcircumpolar.com.

Perhaps the main issue that continues to threaten the conclusion of the convention is funding to assist developing nations in living up to obligations they will soon assume. To date, only Canada has announced money for this purpose. ICC Canada and other coalition members pressed the federal government to make a financial commitment to the convention in the February budget speech and subsequently applauded the announcement by the Minister of Finance.

Developed nations generally prefer the Global Environment Facility (GEF) as the convention implementation financing instrument. The developing world disagrees, claiming that the GEF is difficult to access, overly bureaucratic, less than timely and poorly financed. Instead, the developing world insists upon the establishment of a new fund through the convention to which the developing world would have easier access. The multi-lateral fund established through the Montreal Protocol is seen as a useful model. John Buccini, a civil servant in the Canadian federal Department of the Environment, chairs the international negotiations, including an intersessional process on financing the convention. The coalition continues to seek ways and means to support his efforts.

Aboriginal partners organizations are committed to explaining the results of research conducted under the auspices of the NCP. The same principle applies to our joint involvement in international negotiations. All members of the coalition explain to their constituents and to local and regional media the importance of international negotiations and the utility of a global POPs convention to "solve" this problem over the long term. We have been successful in placing major stories and opinion editorials in Canadian national newspapers. ICC Canada recently unveiled a new Web site, www.inuitcircumpolar.com, which features our work on POPs. In particular, this Web site includes *Silarjualiriniq*, the quarterly journal of ICC Canada, the second issue of which deals exclusively with POPs. As well, we have successfully presented ourselves to the international media and have generated scores of stories on this level. Klaus Topfer, Executive Director of the United Nations Environment Programme, now characterizes Arctic Indigenous peoples as the "conscience" of the negotiations.

The coalition cooperates with several environmental and public interest organizations, including the World Wide Fund for Nature, the Indigenous Environmental Network, the International POPs Elimination Network and the Canadian Arctic Resources Committee to publicize our views. During the Bonn negotiations, both ICC Canada and the CYFN introduced traditional dance troops and traditional/country food to the international audience in order to help them understand the cultural dimension of this environmental and public health issue. Funding for this was provided by the Government of Germany and The Salamander Foundation.

Much of CAIPAP's work is directed to the Government of Canada. The coalition aims to persuade the federal government to take a position internationally that will protect our health and lead to the elimination of offending POPs. To do this requires coalition members to not only meet with civil servants and to deal with the media, but to press parliamentarians. In the last year, CAIPAP has intervened before parliamentary committees examining the *Canadian Environmental Protection Act* and potential amendments to the *Pest Control Products Act*, and participated in the Parliamentary EcoSummit. On these occasions the coalition illustrated the northern and international dimensions to the POPs issue and the need for strong domestic legislation and policy if Canada is to effectively advocate internationally on behalf of northern Indigenous peoples. Advice from CAIPAP is well reflected in the reports of the Standing Committee of the House of Commons on Environment and Sustainable Development. As well, the coalition has given three informal briefings to liberal members of the House of Commons on POPs issues.

Toward the end of the 1998-1999 fiscal year, the coalition presented information to the Joint Public Advisory Committee of the North American Council on Environmental Co-operation (CEC) when this body met in Anchorage, Alaska. Partly as a result of this, a computer modeling study of dioxin transfers from the USA to Nunavut was initiated by the CEC. The coalition advisor and staff from ICC Canada were formal advisors to this work throughout the 1999-2000 fiscal year. The report was unveiled in the June 2000 meeting of the CEC in Dallas, Texas.

ICC is a "permanent participant" in the eight-nation Arctic Council. This forum, including working groups that report to the Council, have been used by ICC Canada to generate interest in global POPs negotiations and to promote ratification by the Arctic states of the POPs protocol to the 1979 UN/ECE Convention on Long-range Transboundary Air Pollution. The council action plan on pollution stresses the need for Arctic states to work together in international fora when Arctic interests are at stake. This plan is to be approved by ministers in October 2000. CAIPAP believes that the ongoing international POPs negotiations represent an immediate test of this principle, and will continue to press Arctic states to advocate for a global POPs convention that protects Arctic Indigenous peoples. The Gwich'in

Steering Committee is likely to become a permanent participant to the Council to be followed by an umbrella organization that links together the Dene Nation, Métis Nation-Northwest Territories, CYFN, and Athabaskan peoples in Alaska. These political developments would greatly strengthen the ability of the coalition to work together on contaminant-related issues.

Expected Project Completion Date

Ongoing

Facilitation of International Action Related to Long-Range Transport of Contaminants into the Arctic

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Northern Contaminants Program Management Committee

Abstract

Persistent organic pollutants (POPs) from distant sources are transported mainly via the atmosphere to the Arctic where they accumulate in humans and in the Arctic food chain. The Northern Contaminants Program (NCP) supports the development of international controls on the use of these substances of concern. The support for these controls includes the following four initiatives: i) the POPs protocol under the United Nations Economic Commission for Europe Convention on Long-Range Transboundary Air Pollution, for which work is underway to ensure compatibility of contaminant trends and survey activities under the NCP with trends and survey information gathering under the Convention; ii) facilitating negotiation of global actions on POPs, for which Canada has contributed $20 million for capacity building in developing countries in support of these negotiations; iii) the Arctic Monitoring and Assessment Programme (AMAP) which monitors the levels of, and assesses the effects of, anthropogenic pollutants in the circumpolar Arctic environment, and for which the NCP has focussed on implementing the Canadian contribution to the second phase of AMAP; and iv) the Canada-Russia Program on Scientific and Technical Cooperation in the Arctic and the North which gathers information on transport pathways of certain contaminants from Russia into the Arctic, and for which an inventory of PCB use and stockpiles in western Russia has been completed.

Objectives

1. To facilitate international cooperation to enable the identification of the significance of long-range contaminant sources outside of Canada and their transport pathways.
2. To develop and maintain appropriate international controls on emissions and discharges.

Introduction

Persistent organic pollutants (POPs) from distant sources are transported to the Arctic mainly via the atmosphere.

They accumulate in humans and in the Arctic food chain and reach levels of concern in many traditional food species important to northern Aboriginal people. The Northern Contaminants Program (NCP) was established to define the significance of observed levels, to provide information to Northerners on possible dietary health implications, and to support the development of appropriate international controls on the substances of concern.

The reduction and elimination of contaminant input to the Arctic requires comprehensive international cooperation and includes the following elements:

- Maintaining the legally binding protocol on POPs negotiated under the Convention on Long-Range Transboundary Air Pollution (LRTAP) to apply in the region of the United Nations Economic Commission for Europe (UN ECE). This region covers much of the northern hemisphere.
- Facilitating and negotiating global actions on POPs to be applied by nations from outside of the geographic area defined by the LRTAP Convention;
- Support for the Arctic Monitoring and Assessment Programme (AMAP), whose goal is to monitor the levels of, and assess the effects of, anthropogenic pollutants on all components of the circumpolar Arctic environment; and
- Support for the Canada-Russia Program on Scientific and Technical Cooperation in the Arctic and the North. An activity has been established under this program to gather information from the Russian segment of the Arctic to understand the transportation pathways of certain contaminants from Russia into the Arctic.

Activities in 1999-2000

UN ECE LRTAP Convention

The legally binding LRTAP protocol on POPs was signed by 36 countries (including Canada) at the UN ECE Ministerial Conference in Aarhus, Denmark in 1998. The agreement will come into force as a legally binding document for those countries that have ratified it when it has collected 16 ratifications. As of June 2000, five countries have ratified it. The agreement bans or severely restricts the manufacture, use or loss to the environment of initially 16 substances, including all POPs of concern in the Canadian Arctic. It also includes a mechanism allowing for the later addition of more substances. A similar protocol controls the manufacture, use or loss of the heavy metals cadmium, lead and mercury. Work activities in 1999-2000 related to preparing to meet the obligations of the protocol once it has entered into force.

Development of action on POPs in non-UN ECE regions

In Montreal in June 1998, Canada hosted the first negotiating session to achieve global controls on POPs through a new Convention under the auspices of the United Nations Environment Programme (UNEP). All of the draft agreement has now been subject to negotiation. The next (fifth) negotiating session (INC-5) is scheduled for the week of December 3, 2000, in Johannesburg, South Africa. It is anticipated that this will be the final session, enabling the Diplomatic Conference to occur in Stockholm in May 2001 for adoption of the Convention. Key issues in the negotiations have included:

- the nature, extent and management of technical assistance which will be made available to developing countries as they move to eliminate their use of POPs,
- the possibility of differential obligations allowing certain developing countries to phase out their use of POPs over a longer time frame than will be allowed for developed countries,
- the need to gather information on levels of POPs in developing countries to convince them that to control POPs should be a domestic priority for their governments, and
- whether it will be necessary to include trade restrictive measures if both production and use of POPs is eliminated.

AMAP

At the inaugural 1991 Rovaniemi Ministerial Conference under the Arctic Environmental Protection Strategy (AEPS), it was agreed to develop an Arctic Monitoring and Assessment Programme (AMAP) "*to monitor the levels of, and to assess the effects of anthropogenic pollutants in all relevant components of the Arctic environment*". The first AMAP assessment included considerable contributions from the NCP and was presented both at the June 1997 Alta, Norway, AEPS Ministerial conference, and at the September 1998 Iqaluit Arctic Council Ministerial meeting. At Alta, Ministers provided guidance on future activities for AMAP. The key features relating to POPs, heavy metals, and radionuclides included: i) increased emphasis on effects (including combined effects) and on human health implications; and ii) time trend studies at key locations.

Activities in 1999-2000 focussed on implementing the Canadian contribution to the second phase of AMAP and providing our share of the support to the AMAP infrastructure. Canada, largely through NCP funds, continued to support the archived freshwater data accessible for the second AMAP assessment at the Freshwater Institute in Winnipeg, Manitoba, and sponsored the AMAP Working Group meeting in Toronto in November 1999.

Canada-Russia Program on Scientific and Technical Cooperation in the Arctic and the North

At the conclusion of negotiations of the LRTAP POPs Protocol, Russia stated that its dependence on PCBs for electrical transmission purposes precludes their commitment to a ban on production of these substances and that therefore they were unable to sign and ratify the agreement. In 1998, the NCP began contributing to an AMAP-led project to assist Russia with this issue. The initial phase of this project consists of compiling an inventory of PCB use and stockpiles in western Russia.

In addition, the NCP, the Canadian International Development Agency and AMAP have cooperated to establish a POPs atmospheric monitoring facility at Amderma in Russia's western Arctic region. The station has an operation time frame of May 1999 to September 2001 for POPs analysis and utilizes identical equipment to that employed at Alert, Nunavut. In 2000, equipment needed to measure total gaseous mercury in ambient air was also installed at this station using NCP funding. Mercury and POPs data from this air monitoring station will be available to NCP and AMAP scientists for inclusion in upcoming assessment reports.

Results and Discussion

UN ECE LRTAP Convention

The LRTAP Convention Working Group on effects is developing plans for trend and survey information gathering, particularly in the area of environmental monitoring, to support the POPs protocol. Canada and the NCP are working to ensure that NCP trend and survey activities which may comprise a substantial proportion of Canada's contribution are compatible with these emerging plans. More information on the LRTAP protocol on POPs can be found at the UN ECE Web site: http://www.unece.org/env.

Development of action on POPs in non-UN ECE regions

Moving the agreement on POPs in non-UN ECE regions into action will present a very difficult challenge for most developing countries where POPs are still heavily utilized. Canada announced at INC-4 a contribution of $20 million (Canadian) for capacity building in support of the agreement. We are the only country to have made a specific commitment of this nature.

The NCP has assisted the negotiations by:

- making available to the negotiators information on long-range transport of POPs and on exposure to wildlife and humans in the Arctic,
- supporting the Canadian Arctic Indigenous Peoples Against POPs (CAIPAP), which has been very effective at explaining northern issues at the negotiations,
- developing the concept of a global POPs monitoring program which would be incorporated into the convention as a tool to evaluate its effectiveness, and
- supporting capacity building in developing countries in their programs to move away from the use of POPs. Southeast Asia has been targeted since this region is believed to be a significant source of POPs emissions and discharges. In this period, a training session on Integrated Pest Management was co-hosted with UNEP in Thailand. This initiative has confirmed that the challenge of achieving global controls on POPs will be to ensure that developing countries have the capacity to comply with any future POPs actions.

More information on the POPs negotiations under the UNEP can be found at the UNEP Web site: http://www.unep.ch.

AMAP

The next major AMAP assessment will consist of a series of reports on individual topics (e.g. POPs, heavy metals, radionuclides, human health, etc). Recalling the presentation of the first AMAP report to the UN Commission on Sustainable Development (CSD) in 1997, and the desire of Ministers expressed at Alta to bring the attention of the world to the state of pollution of the Arctic environment, the next assessment will be completed in time for a possible Rio+10 meeting under the CSD in 2002. A "progress report" from AMAP will be available for ministers at their conference in Point Barrow, Alaska in October 2000.

More information on AMAP activities can be found at http://www.amap.no.

Canada-Russia Program on Scientific and Technical Cooperation in the Arctic and the North

The initial phase of the AMAP-led project to assist Russia in signing and ratifying the LRTAP POPs Protocol has been completed. This phase consists of compiling an inventory of PCB use and stockpiles in western Russia. This work provides the basis for the development of preventative and remedial action plans and identification and utilization of alternatives to PCBs.

Conclusions

Work under the above initiatives is continuing.

Expected Project Completion Date

Ongoing

Program Management

Northwest Territories Environmental Contaminants Committee

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Abstract

The Northwest Territories Environmental Contaminants Committee (NWTECC) membership is composed of representatives from various departments of the federal and territorial governments as well as national and regional Aboriginal partners. The Committee meets regularly throughout the year to facilitate scientific study and assessments, as well as the communication of information to Northerners on the presence and possible effects of contaminants in the environment. The NWTECC also facilitates the communication of northern priorities to researchers.

Objectives

1. To provide a forum for the two-way transfer of contaminants information between Northwest Territories (NWT) Northerners, researchers, the Northern Contaminants Program (NCP) and other contaminants-related programs.
2. To establish a communications network that ensures Northerners are informed and involved in contaminant-related activities.
3. To identify priorities and information gaps related to environmental contaminants research in the NWT.
4. To act as a central repository for environmental contaminants information.

Table 1. Meetings held by the Northwest Territories Environmental Contaminants Committee during the 1999-2000 fiscal year

Meeting Name	Meeting Date	Meeting Type	Key Agenda Items
NWTECC7	April 8, 1999	Conference call	Election of Chair and Vice-Chair Roundtable identification of issues NCP – review LCC proposals; Management Committee meeting
NWTECC8	May 6, 1999	Conference call	NCP– review of outstanding LCC proposals Roundtable identification of contaminant concerns
NWTECC9	June 25, 1999	Conference call	Roundtable identification of issues Update from GNWT Health and Social Services on Health Committee and Advisories
NWTECC10	July 23, 1999	Conference call	Review of Monitoring, Health and Communications Blueprints Roundtable identification of issues Update on GNWT Activities
NWTECC11	September 8, 1999	Conference call	Roundtable identification of issues Selection of five representatives to attend NCP Results Workshop Update from GNWT Health
NWTECC12	November 19, 1999	Conference call	NCP 2000-2001 Call for Proposals NCP Reports Roundtable identification of issues
NWTECC13	There was no meeting	Conference call	
NWTECC14	January 13, 2000	Conference call	NCP Call for Proposals Fisheries Advisories Guide Review of Local Contaminants Concern Proposal Roundtable discussion of contaminants-related activities
NWTECC15	February 3 & 4, 2000	In-person meeting in Yellowknife	Review of NCP Blueprints and criteria to be used Review of NCP Proposals that relate to the NWT Roundtable discussion of contaminants-related activities

5. To provide advice on appropriate funding sources.
6. To review NWT proposals for the NCP prior to full technical reviews.

Introduction

The NCP is now in its second phase, with more than eight years of research to date. The program has evolved to include NWT communities and regions extensively in research and communication efforts. This has increased the need to have a central body that can coordinate contaminants information and research initiatives. The Northwest Territories Environmental Contaminants Committee (NWTECC) provides an opportunity for researchers and communities to provide input into contaminants activities and express their concerns.

The membership is composed of representatives from various departments of the federal and territorial governments as well as national and regional Aboriginal partners, making it particularly suitable for addressing NWT research priorities and information gaps.

Activities in 1999-2000

The NWTECC met eight times to discuss various contaminants related issues (Table 1). Frequent roundtable discussions and updates helped to keep contaminants workers in governments and regional and territorial Aboriginal organizations informed of study results, current activities, upcoming proposals or work and conferences. In addition, to provide an opportunity for committee members to become better informed of recent study results, the NWTECC sent five representatives to attend the annual NCP Results Workshop in White Rock, British Columbia, in September1999.

The NWTECC held a two-day in-person meeting in February 2000 to review the NCP proposals of immediate relevance to the NWT. Prior to the proposal review, the monitoring, health and communication blueprints were reviewed by the Committee. The Committee also considered Local Contaminants Concerns proposals from communities and approved five for funding in 1999-2000.

During 1999-2000, the Committee underwent reorganization to reflect the creation of Nunavut and the new, smaller Northwest Territories. Work to develop a communications network was ongoing this year, as were efforts to expand a resource library.

Results

Notes that summarize the activities and decisions of the Committee from each of the eight meetings are available from the Contaminants Division, Department of Indian Affairs and Northern Development in Yellowknife.

Discussion and Conclusions

The NWTECC has proved to be an effective forum for discussing contaminants-related concerns and for providing northern input to the NCP.

Expected Project Completion Date

The NWTECC expects to continue operating through the 2000-2001 fiscal year and beyond.

The Yukon Contaminants Committee Communications Program

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Abstract

The Yukon Contaminants Committee (YCC) coordinates Northern Contaminants Program (NCP) work in the Yukon. The committee consists of representatives from the federal government, the Yukon Territorial Government, Council of Yukon First Nations (CYFN), Yukon Conservation Society, Environment Canada, Department of Fisheries and Oceans and Yukon College. The YCC's role, in addition to representing Yukon interests in the NCP, is to act as a liaison between researchers and Northerners. The YCC meets at least twice every year and as required. It coordinates Yukon studies under the NCP. The Department of Indian Affairs and Northern Development chairs the YCC and provides administrative support and, in partnership with CYFN, drafts most proposals for submission to the NCP. The types of studies carried out in the Yukon are diverse and include the Traditional Foods Monitoring Program, curriculum development, traditional knowledge guidelines, fact sheets, local source contaminants of concern, cancer registry, community and territorial workshops, and community training.

Objectives

1. To answer Northerners questions concerning contaminants issues.
2. To communicate the results of work carried out under the Northern Contaminants Program (NCP) to all Yukon residents.
3. To act as a link between researchers and stakeholders.
4. To educate Northerners about contaminant issues.

Introduction

The Yukon Contaminants Committee (YCC) was established in 1992 to address contaminants concerns within the Yukon. The YCC acts as a link between the scientific community and Northerners. The core membership is diverse and consists of representatives from several federal and territorial departments, Council of Yukon First Nations, Yukon Conservation Society and Yukon College. Several individual First Nations have also played a key role on the Committee.

The YCC coordinates the projects funded under the NCP occurring in the Yukon in order to reduce overlap and to allow better coordination between researchers and Northerners. The majority of the budget received for the operation of the Committee covers travel costs to attend meetings and workshops in the Yukon and across Canada.

The YCC has a responsibility to oversee all studies carried out under the Local Contaminants Concerns Program (LCC). All studies are approved under the LCC and the LCC ensures that results are reported back to the NCP.

One of the largest challenges facing the YCC is the education and communication of scientific information to Northerners. Prior to the release of any scientific information it is essential to provide the public with the capacity to fully understand the information. In an attempt to accomplish this the YCC has coordinated several information and education programs under the NCP.

Activities in 1999-2000

The YCC met three times as a whole and sub-groups met as required to deal with specific issues. The chair of the YCC and the Yukon Territorial Government representatives attended the NCP management committee meetings to represent YCC interests. The committee reviewed Yukon proposals for the social-cultural review process as well as reviewed and approved all Yukon education and communications proposals. The Committee funded two representatives to attend the results workshops. Members also attended community meetings and workshops as requested during the year.

Results

All YCC activities are reported to the NCP by the chair to the management committee at the semi-annual NCP management committee meetings.

Discussion and Conclusions

The YCC will continue to be the primary contact for contaminants concerns in the Yukon.

Expected Project Completion Date

The YCC will be requesting continuing funding for the duration of the NCP.

Development of the Nunavut Environmental Contaminants Committee

- **Project Leaders:**

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- **Project Team:**

  Members of the interim NECC, including representatives from: Inuit Tapirisat of Canada; Nunavut Tunngavik Incorporated; Environment Canada; Department of Fisheries and Oceans; Kivalliq Inuit Association; Kivalliq Wildlife Federation; Qikitaaluk Wildlife Board; Qitiqtani Inuit Association; Government of Nunavut Department of Health and Social Services; Government of Nunavut Department of Sustainable Development; Government of Nunavut Water Board

Abstract

The Nunavut Environmental Contaminants Committee (NECC) is a newly formed committee dealing with contaminants projects in Nunavut. Membership is growing, and currently consists of representatives from various departments of the federal and territorial governments as well as national and regional Aboriginal partners. The Committee meets regularly throughout the year to facilitate scientific study and assessments, the communication of information to Northerners on the presence and possible effects of contaminants in the environment, and the communication of Northern priorities to researchers. The NECC assists in the process of collaborative study, assessment and communication of information to Nunavut residents about the presence and possible effects of contaminants in the air, land, water and wildlife and humans by, for example: i) providing recommendations on contaminants research priorities for the proposals submitted to the Northern Contaminants Program; ii) establishing working groups to deal with specific issues, such as contaminant sources within Nunavut; and iii) funding local contaminant concerns studies, such as the current site assessment of Nottingham Island.

Objectives

1. To provide a forum for the two-way transfer of contaminants information between Nunavut residents, researchers, the Northern Contaminants Program (NCP) and other contaminants-related initiatives.
2. To facilitate the incorporation of Inuit Qaujimajatuqangit into the study, assessment and communication of environmental contaminants information, concerns and research priorities.

3. To facilitate a communications network that ensures Nunavut residents are informed of, and involved in, contaminant related activities.
4. To identify community level priorities and information gaps related to environmental contaminants in Nunavut.
5. To provide updates on research activities in Nunavut that relate to contaminants in the environment, wildlife and people.
6. To act as a clearing house for contaminants information to assist Nunavut residents in making informed decisions about the harvesting and consumption of traditional foods.
7. To develop communication strategies to effectively relay information regarding contaminants.
8. To provide advice to communities on appropriate funding sources for contaminants research and remediation projects.
9. To review NCP and other Nunavut related proposals in accordance with the mandate and priorities of the Committee.

Introduction

Contaminant and contaminants-related research has been taking place under the auspices of the NCP since 1991. The program has evolved to include all northern communities and regions extensively in research and communication efforts. This has increased the need to have a central body in each territory that can coordinate contaminants information and research initiatives within their respective regions.

Nunavut was established in April 1999, and thus there was a need to establish a contaminants committee in the new territory to reflect the different contaminant issues faced by the people of Nunavut. Much of the work in 1999-2000 consisted of getting the Nunavut Environmental Contaminants Committee (NECC) up and running, designing a terms of reference and making the shift from an interim committee to a formal one. Securing participation and representation from a broad variety of organizations has been a primary challenge. By establishing the NECC, it is hoped that it will provide a one-window opportunity for researchers and communities to provide input into contaminants activities and to express their concerns.

The membership of the NECC is composed of representatives from various departments of the federal and territorial governments as well as national and regional Aboriginal partners, making it particularly suitable for addressing Nunavut research priorities and information gaps.

Activities in 1999-2000

The interim NECC met formally four times to discuss various contaminants related issues (Table 1) including one two-day in-person meeting to review the NCP proposals of relevance to Nunavut. There were also numerous conversations and e-mails throughout the year.

Results

Minutes from the NECC's four meetings in 1999-2000, summarizing the activities and decisions of the Committee, are available in both English and Inuktitut from the Chair of the NECC. Also available are the terms of reference for the Committee.

Major decisions of the NECC included recommendations on NCP funding proposals. Recent achievements include the funding of a Local Contaminants Concerns fund project out of Coral Harbour, the finalization of the terms of reference, increasing participation from Inuit organizations, and the establishment of a working group to investigate locally generated dioxins and furans in Nunavut. Other activities have included the circulation of information and discussions surrounding new contaminants issues as they are identified, and the production of public information relating to the NECC.

Discussion and Conclusions

The interim NECC has proved to be an effective forum for discussing contaminants related concerns and for providing northern input to the NCP during its development and it is expected to continue this kind of work in the current fiscal year.

Expected Project Completion Date

The NECC will continue operating through the 2000-2001 fiscal year.

Table 1. Meetings held by the interim Nunavut Environmental Contaminants Committee during the 1999-2000 fiscal year

Meeting Name	Meeting Date	Meeting Type	Key Agenda Items
NECCi1	September 29-30, 1999	In-person meeting in White Rock, BC	NCP Overview NECC Goals and Terms of Reference
NECCi2	December 13, 1999	Conference call	Proposal Review Process Preparation for NECC Phase II
NECCi3	February 15-16, 2000	In person meeting in Iqaluit	NCP Review Process NCP Project Review – Nunavut
NECCi4	March 21, 2000	Conference call	Discussion – Proposal Review Kitikmeot Contaminants Committee Proposal Advisory on Consumption of Glaucus Gull Eggs

Northern Contaminants Program Interlaboratory Quality Assurance Program for 1999-2000

◆ **Project Leader:**

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◆ **Project Team:**

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Abstract

This report describes the quality assurance (QA) activities conducted during 1999-2000 in support of the Northern Contaminants Program (NCP). Interlaboratory assessments included a study on the analysis of heavy metals and methylmercury in fish and seal muscle tissues, and another on organochlorines and polychlorinated biphenyls (PCB) congeners in fish, mussel homogenate and injection-ready solutions. The development of a "minimum list" of 30 PCB congeners for NCP analyses is presented and compared to the target PCB congeners of other international research, monitoring and QA programs. Efforts were also focussed on designing a series of toxaphene intercomparisons that would include all NCP laboratories, from those that analyze for toxaphene in air or water samples, to those analyzing fish, mammals and human tissue samples. In addition, this report summarizes the target analytes and matrices under study in the NCP-funded research projects for 1999-2000, along with an evaluation of the data quality needs for radionuclide measurements. After updating the survey information on the laboratories contributing measurement data to the NCP, recommendations for participation in specific external intercomparison programs were made for toxaphene, radionuclides and brominated diphenyl ethers.

Objectives

The main objective of the Northern Contaminants Program (NCP) Quality Assurance (QA) Program is to provide information on the overall quality of the NCP's measurement data to its management committee in order that they may make informed decisions on the sources and effects of contaminants on the Arctic environment and on human health. As a result, these decision makers would be assured that their contributions toward the establishment of international agreements and controls to protect the health of the Arctic ecosystem and its inhabitants are based on a scientifically sound database of information.

A second objective is to provide the research managers of NCP-funded studies with assurance of the quality, reliability and comparability of measurement results produced by laboratories contributing data to their research. This is primarily achieved by conducting intercomparison exercises on various contaminants of concern, such as persistent organic pollutants (POPs), radionuclides and heavy metals.

Specifically, the goals for 1999-2000 were to:

1. Maintain an inventory of measurement capabilities and interests among the NCP laboratories in order to provide advice to the QA Technical sub-Committee on the status of the NCP's QA needs and on emerging issues (e.g. new contaminants of concern) and changes in priority.
2. Propose, coordinate and review participation by NCP laboratories in external national and international intercomparison exercises, where such participation would be considered to be more cost-effective and would add value to this QA Program.
3. Investigate and report on any specific QA needs for radionuclide measurements in the NCP research studies and propose on how to best fulfill these needs.
4. Develop a list of PCB congeners that should be analyzed for all NCP-funded work and compare this list with similar approaches made for the Arctic Monitoring and Assessment Programme (AMAP) and for the International Atmospheric Deposition Network (IADN).
5. Conduct an interlaboratory study on the analysis of heavy metals including mercury, methylmercury, arsenic and selenium in fish and marine mammal tissue.
6. Conduct an interlaboratory study on the analysis of organochlorine pesticides and PCBs in standard solutions and biota.
7. Review and compare the methodologies and quantitation techniques used for toxaphene analysis by NCP laboratories and make recommendations for participation in a suitable external intercomparison study, if available.

Introduction

The NCP, like all research and monitoring programs, requires an on going QA program that provides assurance to its managers of the quality and comparability of measurement results being generated for their research projects. At the same time, it should meet the diverse QA/Quality Control needs of the researchers and analysts by providing them with appropriate diagnostic tools for their analyses and offering guidance and support toward corrective measures, if needed. The design of this QA program also aims to identify sources of measurement uncertainties and variation of analytical results in order to provide information on data quality and reliability to the management committee of the NCP.

The main activities are the intercomparison exercises, which focus on contaminants of concern to Canada's Northerners. Participation in these studies ensures that laboratories conducting measurements for the NCP are producing reliable data with acceptable levels of precision and accuracy, and ensures comparability among the various research and analytical laboratories and between the NCP research projects. In addition, by serving as a diagnostic tool for the participating laboratories, these studies provide a means for continual improvement in the measurement process.

The submitted laboratory data for both standard solutions and matrix test samples are evaluated for accuracy and comparability by Z-scores and by comparison with the target concentrations, for precision on replicate analyses (or by percent difference on blind duplicate samples), and for bias by a modified Youden ranking procedure. The data assessment is then used as a diagnostic tool for the participants to apply corrective action, as needed. In addition, where possible, the data set for the NCP QA studies is compared to the results of other similar external intercomparisons. Thus, the complete study report provides a snapshot of data quality to the management committee of the NCP along with an overview of the capabilities and comparability of the NCP laboratories conducting these measurements.

Activities in 1999-2000

The first year of the QA program of Phase II of the NCP began with a survey of the project leaders and the laboratories conducting NCP-funded analyses (Stokker and Gomes 1998). This information is updated annually in order to maintain a priority list of the analytes and relevant matrices for the intercomparison studies being conducted. For 1999-2000, these were determined to be heavy metals, methylmercury, toxaphene, organochlorines and PCBs in biotic samples. In addition, an assessment of the QA needs for radionuclides within the NCP led to the conclusion that participation in external interlaboratory programs would prove to be a more cost-effective means for assuring the data quality of NCP radionuclide measurements.

The second survey of external national and international QA programs conducted in 1998-1999 identified several external interlaboratory programs that would complement those run in the NCP QA program (Stokker and Gomes 1999). Specific recommendations were made for participation in two external interlaboratory studies in 1999-2000. The first was a development exercise for toxaphene, conducted by the Quality Assurance for Marine Environmental Monitoring in Europe (QUASIMEME). The second was a brominated diphenyl ether (BDPE) intercomparison study conducted by researchers in the Netherlands. In addition, from the radionuclide measurement review, it became apparent that the QA needs for radionuclide measurements in the NCP studies were being adequately addressed by several appropriate external QA programs. Therefore, it was recommended that continued participation by these laboratories in appropriate intercomparisons conducted by the International Atomic Energy Agency (IAEA) and other agencies would be the most cost-effective means for assuring the data quality of the NCP's radionuclide measurements.

The development of a list of PCB congeners that could be made mandatory for all NCP-funded PCB analyses was completed and forwarded to the QA Technical Sub-Committee for their review. A database on PCB congeners reported in the literature for various marine or Arctic matrices was created and the selection of 30 congeners was made, based on their toxicity, frequency of occurrence and concentration levels relative to the (reported) total PCB levels. This proposed list was then compared with existing PCB congener lists in other external research, monitoring and QA programs.

During 1998-1999, the first intercomparison study for Phase II of the NCP (Interlaboratory Study NCP-II-1) on the analysis for trace metals in sediment was conducted. Eight laboratories provided results and most participants analyzed for at least 10 of the 16 target parameters. The follow-up study in 1999-2000 (Interlaboratory Study NCP II-2) focussed on the analysis of heavy metals and methylmercury in biota. Fifteen laboratories contributed results in this study.

Interlaboratory Study NCP II-3, on the analysis of organochlorines and PCBs in biota, was conducted this year with 14 participants providing data on the test samples.

Finally, a comparison of toxaphene methodologies, calibration standards and quantitation techniques provided the information necessary to design a series of intercomparisons that would assess the quality and comparability of toxaphene measurements for the many different substrates being analyzed within the NCP research projects. These annual intercomparisons will begin in 2000-2001 and will progress from simple test samples and standard solutions through more complex matrices.

Results

Update to the NCP laboratory and research project survey

Table 1 provides a summary of the NCP-funded research projects containing a measurement analysis component. For 1999-2000, 24 research projects involving 17 project leaders are employing the use of 28 measurement facilities. The most common matrices for analysis are fish (10), and marine or terrestrial mammals (nine). Three studies include the analysis of Arctic birds or eggs, and three are investigating human tissue samples (blood, urine or hair). Among the abiotic test samples, five studies include air measurements, five analyze water samples and only two address sediment or soil samples.

The number of NCP-funded studies targeting different inorganic contaminants are: mercury (17), selenium (nine), other toxic heavy metals (12), and methylmercury (six). Only one study proposed analyzing for organotins in 1999-2000, while two facilities are measuring radionuclides. Among the organic parameters of interest, organochlorine pesticides are being measured for 15 research projects, PCBs in 10 studies, toxaphene in seven, and BDPEs in three studies.

External QA programs

Two international intercomparisons were recommended this year to the NCP measurement laboratories. The first was a toxaphene development exercise conducted by QUASIMEME in Scotland. Three NCP laboratories participated in this study which included a series of standard solutions and cleaned-up fish tissue extracts to test the methods of separation and quantification of the toxaphene congeners CHB26, CHB32, CHB40, CHB41, CHB44, CHB50 and CHB62.

Two laboratories are currently part of a BDPE study being conducted by researchers in the Netherlands in collaboration with the Bromine Science and Environmental Forum. The analysis of three mandatory and eleven optional BDPE congeners were requested in five biota and two sediment test samples.

Several of the NCP laboratories participate in a variety of other external QA programs on an ongoing basis. The two facilities conducting radionuclide measurements for the NCP continue to participate in the IAEA intercomparison exercises for radionuclides, when available (see discussion below). In addition, many of the NCP measurement laboratories are accredited by and participate in the

Table 1. Target measurement analyses for Northern Contaminants Program projects in 1999-2000

Project Leader	Agency*	Lab analyses	Matrix	Hg	Se	Other TMs	MeHg	RNs	Stable isotopes	OCs/POPs	PCBs	Toxaphene	Other parameters
Barrie	MSC	√	air							•	⊗	⊗	PAHs
Bidleman	MSC	√	air, water							chlordanes	coplanars	•	PCNs, BDPEs, CPs, chloroparaffins
Schroeder	MSC	√	air, snow	•									TGM
Poissant	MSC	√	air, snow, soil	•									in situ TGM, ozone in air
Chan	CINE	(effects)	rats				⊗						
		√	seal	•	•								
		(model)	fish, mammals							⊗	⊗	⊗	
		√	fish, mammals	•						•	•	•	
Ayotte	CHUQ	(effects)	infants	⊗						⊗			
		√	hair, blood	•	•		•						
		(effects)	children				⊗				⊗		
		√	blood			•	•			•	•		
Lockhart	FWI	√	fish	•	•	As, Pb, Mn	•		•				
		√	sediment			Pb		•	•	•			(PAHs)?, Pb^{210}, Cs^{137}
Stern	FWI	√	burbot	•	•	As				•	•	•	coplanar PCBs, PBBs, BDPEs, CDPEs
		√	beluga										PBBs, BDPEs, CDPEs
Elkin	GNWT	√	caribou	•		•		•					Cs^{137}, K^{40}, Sr, Po^{210}
MacNeil	IRHSSB	√	hair, blood	•		•	•			•			
Evans	NWRI	√	fish, water	•		•			C,N	•			
		√	fish	•	•	As			C,N	•	•	•	coplanar PCBs
Braune	NWRC	√	seabirds	•	•				•	•	•		stable isotope N^{15}/N^{14}
		√	oldsquaw ducks	•	•	Cd, Pb, Cu			C,N,S				
Norstrom	NWRC	√	polar bears							•	•		OH-PCBs, retinol
Muir	NWRI	√	fish/shellfish, mammals	•		•	•			•	•	•	coplanars, organotins
		√	char, mussels	•	•	Pb, Cd, Zn				•		•	
		√	biota, air, water							HCHs			

Canadian Association for Environmental Analytical Laboratories (Ottawa, ON) environmental performance evaluation studies.

Radionuclide review

Only two research projects incorporated the analysis of radionuclides in their work this year. These measurements are being conducted at two different facilities, both of whom routinely participate in more than one international program of radionuclide intercomparisons. The first facility has a full QA program in place in compliance with the Atomic Energy Control Board requirements and ISO Guide 25. They also participate regularly in interlaboratory programs conducted by the US Environmental Protection Agency (EPA) National Exposure Research Laboratory (NERL), by Chalk River Labs of Atomic Energy of Canada Limited, and by the World Health Organization's International Reference Centre for Radioactivity in France. The second institution participates frequently in the intercomparisons offered by the US-EPA NERL and by the IAEA in Austria.

Development of a list of "mandatory" PCB congeners

Polychlorinated biphenyls (PCBs) are ubiquitous environmental pollutants that have been measured and quantified in a wide variety of matrices by countless different techniques over many years. Despite the recent improvements in available standards and in methodologies, it remains difficult to compare results generated by reference to Aroclors or to "total PCBs." For the NCP, the concentrations of various PCB congeners have been and continue to be measured in diverse matrix materials such as air, water, sediment, fish, birds, mammals, and human tissue. However, the selection of congeners measured by each analyst has often been selected on the basis of matrix expectations and historical results, and has therefore been uniquely different for each laboratory in the program. In order to compare data between laboratories over time and between sample types, it became necessary to develop a minimum or "mandatory" list of congeners, essential to the NCP for comparative purposes.

A literature search for PCB congener concentrations in marine or Arctic samples was conducted in order to tabulate the reported levels for various matrices: water, air/snow/rain, vegetation, sediment, fish, mussels, mammalian and bird tissues, blood (human and seal), and other human tissue samples. From these data, the selection of congeners to be reported in NCP work was made, based on their toxicity, frequency of occurrence and concentration levels relative to the (reported) total PCB levels.

Table 1. Target measurement analyses for Northern Contaminants Program projects in 1999-2000

Project Leader	Agency*	Lab analyses	Matrix	Hg	Se	Other TMs	MeHg	RNs	Stable isotopes	OCs/POPs	PCBs	Toxaphene	Other parameters
Wayland	WQL-PNR	√	eiders	•	•	Cd	•						
Palmer	YCC	√	fish							•	•	•	
Feeley	HC-BCS	NO	standards								•		risk assessment of dioxin-like PCBs
Total		**24**	**air (5), water (5), sediment/soil (2), fish (10), birds (3), mammal (9), human tissue/blood (3)**	**17**	**9**	**12**	**6**	**2**	**6**	**15**	**10**	**7**	**** 28 different analytical locations****

* MSC, Meteorological Service of Canada; CINE, Centre for Indigenous Peoples' Nutrition and Environment; CHUQ, Centre Hospitalier Universitaire de Québec; FWI, Freshwater Institute; GNWT, Government of the Northwest Territories; IRHSSB, Inuvik Region Health and Social Services Board; NWRI, National Water Research Institute; NWRC, National Wildlife Research Centre; WQL-PNR, Water Quality Laboratory - Prairie and Northern Region; YCC, Yukon Contaminants Committee, HC-BCS, Health Canada - Bureau of Chemical Safety

• measurement analyses conducted

⊗ data analysis or toxicological effects only; little or no measurements anticipated during 1999-2000

The proposed NCP list of 30 congeners (Table 2) includes the "ICES 7" (the seven congeners deemed essential by the International Council for the Exploration of the Seas) and the congeners most commonly targeted by other international agencies and programs such as AMAP (Janine Murray, personal communication), IADN (Hoff 1999), National Oceanic and Atmospheric Administration (NOAA) (National Oceanic and Atmospheric Administration 1993), GULFWATCH (Chase et al. 1998), QUASIMEME (De Boer and Wells 1997), the Quebec Ministry of Environment (Lévesque and Moore 1998), and the Canadian Shellfish Guidelines (Dumouchel and Hennigar 1995). Furthermore, because this target mix of selected congeners includes those evaluated in other QA Programs, comparisons of the NCP measurements with those of other international research and monitoring programs will be assured for the future.

Table 2. Comparison of Northern Contaminants Program minimum list of PCB congeners with other international programs

PCB Congener No.	1999 NCP Minimum List	AMAP	ICES	IADN	NOAA	GULFWATCH mussels	QUASIMEME	QUEBEC MOE	Canadian Shellfish Guidelines
5		(x)		x		(x)			x
8	√	x/5		x	◆	◆/5			x
15				x		(x)			x
17								x	
18	√	x	x	x	◆	◆/15		x	x
28	√	◆/31	◆	x	◆	◆	x	x	x
29				x		◆			x
31	√	(◆)	x	x			x	x	x
33								x	
44	√	x	x	x	◆	◆		x	x
49	√	x		x				x	
50						◆			
52	√	◆	◆	x	◆	◆	x	x	x
66	√	(x)	x	x	◆/95	◆/95			x
70				x				x	
74	√			x				x	
77	B	x		x	B	◆			x
81	B			x					
82								x	
87	√	x		x		◆		x	x
90		(◆)		x	(x)	(x)			x
95		x/66		x	(x)	(x)		x	x
99	√	x		x				x	
101	√	◆/90	◆	x	◆/90	◆/90	x	x	x
105	√	◆/132	x	x	◆	◆	x	x	x
110	√	x		x				x	x
118	√	x	◆	x	◆	◆	x	x	x
126	B	x		x	B	◆			x
128	√	x		x	◆	◆		x	x
132		(◆)		x		(x)		x	x

Table 2. Comparison of Northern Contaminants Program minimum list of PCB congeners with other international programs

PCB Congener No.	1999 NCP Minimum List	AMAP	ICES	IADN	NOAA	GULFWATCH mussels	QUASIMEME	QUEBEC MOE	Canadian Shellfish Guidelines
138	√	◆/163	◆	x	◆/163/164	◆	x	x	x
146		x							
149	√	x		x				x	
151	√	x		x				x	
153	√	◆	◆	x	◆	◆/132	x	x	x
154						◆			
156	√	x	x	x			x	x	
157		(x)							
158								x	
159					(x)				
163		(x)		x	(x)				x
164					(x)				x
169	B	x		x		◆		x	x
170	√	x	x		◆/190	◆/190		x	x
171								x	
177								x	
178									x
180	√	◆	◆	x	◆	◆	x	x	x
182					(x)				
183	√	x		x				x	
187	√	x	x		◆/182/150	◆		x	x
190					(x)	(x)			x
191								x	
194	√	x		x				x	
195	√	x		x	◆	◆/208		x	x
199								x	
201	√	x/157							
205								x	
206	√	x	x		◆	◆		x	x
208						(x)		x	x
209	√	x	x		◆	◆		x	x
Total # congeners	**30**	**7 essential 39 (total)**	**14-17**	**84**	**18-20**	**25**	**10**	**41**	**35**

Program relevance:

√ - Required for NCP work
◆ - Essential
x - Recommended

B - indicates congeners of toxicological importance
(◆) - co-elutes with an essential congener
(x) - co-elutes with a recommended congener

Interlaboratory Study NCP-II-1

Trace metals in sediment (Stokker and Kaminski, in press)

Eight laboratories provided trace metal results on the four sediment check samples issued in 1998-1999. For most of the metals, the data were quite comparable, as demonstrated by coefficients of variation of less than 25%. The parameters that showed the least comparability were chromium, selenium and vanadium. The repeatability of analysis for each participant was quite good on the blind duplicate samples, with only one laboratory exceeding 10% difference between duplicates for more than one metal. All laboratories exhibited some bias for at least one of the target parameters. Z-scores were satisfactory (i.e. $Z \leq 2$) for all key toxic metals.

Overall, the results of the trace metal in sediment interlaboratory study were in good agreement with previous trace metal assessments conducted by the National Water Research Institute on sediment samples, and are in keeping with assessments reported by QUASIMEME, IAEA and NOAA.

Interlaboratory Study NCP-II-2

Trace metals and methylmercury in biota (Stokker and Kaminski, in press)

Five test samples were shipped in July 1999 to 17 participating laboratories for analyses of heavy metals including arsenic, selenium and mercury, and for methylmercury. The test materials were frozen Tasuijaq lake trout (whole fish), frozen homogenized muscle tissue from Nepihjeer ringed seal, and three freeze-dried certified reference materials (CRMs), including dogfish muscle, cod muscle and mussel homogenate.

Most of the heavy metal results were close to their target values except where the concentration levels were close to the detection limits of the participating laboratories (e.g. arsenic, cadmium and nickel in the lake trout and seal muscle samples). Consequently, there were a considerable number of unsatisfactory Z-scores in this study (i.e. $Z > 3$). However, while there were a number of individual outlying results, only three participants exhibited any metal-specific bias: one was biased low for arsenic, one was biased low for cobalt and a third participant was biased high for cobalt.

There was very good agreement among the participants for their total mercury (15 laboratories) and total organic mercury results (five laboratories). Except for the very low level mussel tissue, the coefficient of variation did not exceed 15% for any of the test samples. Six participants analyzed for the methylmercury species. With the exception of one laboratory (who is not presently reporting heavy metal nor methylmercury data to the NCP) the methylmercury results were excellent in terms of both accuracy (comparison to the CRM reference values) and intercomparability (low coefficient of variation).

Interlaboratory Study NCP-II-3

Organochlorines and PCBs in biota (Stokker and Kaminski, in press)

Among the NCP research studies that analyze for organochlorines and PCBs, the sample matrices for 1999-2000 include air, snow, water, sediment, birds, fish, shellfish, marine and terrestrial mammals, and human blood. Therefore, check samples prepared in solution form (i.e. injection-ready standards) were included in this intercomparison study to facilitate the evaluation of interlaboratory comparability. In addition to the four injection-ready solutions (one for organochlorines only, two for the NCP target list of 30 PCB congeners, and one for the four coplanar PCBs only), the test samples in this study included three naturally contaminated biota samples (two lake trout and one mussel homogenate).

The results provided for the analysis of the injection-ready solutions were generally quite accurate and comparable. This provides considerable confidence in the quality of calibration standards being used by the NCP laboratories. However, the analysis for these same parameters in the tissue samples were considerably less comparable, particularly for the OC measurements. This is in agreement with the findings of other international scientists conducting similar intercomparison studies: that currently available analytical methods for PCBs and organochlorines do not allow for the production of very accurate results when analyte concentrations are < 1 $ng \cdot g^{-1}$ (De Boer and Wells 1997).

For the analysis of PCB congeners in the three tissue samples, the interlaboratory comparability was satisfactory (< 45% coefficient of variation). However, the study results also indicated that data for PCB-66, PCB-95 and PCB-209 are less reliable than those of the other congeners. On a positive note, the data generated for the coplanar PCBs were very good, both in the standard solutions and in the matrix samples.

Toxaphene methodology review

As listed in Table 1, seven NCP research projects proposed toxaphene analyses for 1999-2000. These analyses were to be conducted at four measurement facilities (Freshwater Institute, National Laboratory for Environmental Testing, Centre for Indigenous Peoples' Nutrition and Environment, and AXYS Analytical Laboratories), each of which had previously participated in one or more of the international

QUASIMEME toxaphene development exercises (Stokker and Gomes 1998).

In order to design an intercomparison program that would encompass the needs and capabilities of each of the toxaphene measurement laboratories, a comparison was made of the toxaphene methodologies in use at the above four facilities as well as at those of other Canadian laboratories. From this review, it became clear that negative ion chemical ionization mass spectrometry (NICI-MS) is the predominant technique being used for quantitation. However, electron capture detection (ECD) with confirmation by mass selective detection (MSD), and Ion Trap MS/MS are also being used. Several researchers were quantifying "total toxaphene" against a Hercules technical toxaphene standard, while others employed technical toxaphene standards from the US-EPA or other (commercial) suppliers. Most calculated total toxaphene by using a single response factor against the sum of four to more than 40 peak areas, while two had used multiple response factors for some or all of their total toxaphene calculations. All laboratories surveyed used one or more congener mixtures for the specific analysis of toxaphene congeners, reporting data for Parlar 26 (T2) only, up to more than 20 congener-specific results.

Since the NCP researchers are analyzing a variety of substrates ranging from air samples through water, sediments and various biotic tissues, and because the use of ECD requires the complete removal of interfering compounds such as PCBs and chlordanes, a wide variety of cleanup procedures are currently being used by the different analysts. These differences, therefore, will require that the check samples in each of the intercomparisons include standard solutions in order to facilitate a comparison of the different laboratories and analytical techniques.

From the above information, a series of annual toxaphene intercomparisons have been designed for the NCP QA Program. During the summer of 2000, the first test samples of standards (both technical toxaphene and a congener mix) and a lipid-free burbot liver extract will be issued. Future studies will include samples of increasing complexity, both in terms of the toxaphene components as well as interfering background contaminants. At present, fish samples from Lake Ontario and Lake Superior are being assessed for suitability for use in the toxaphene intercomparison in 2001-2002. To ensure comparability with other international programs that address toxaphene, the selection of target parameters for these studies will incorporate the recommendations put forth by AMAP (Arctic Monitoring and Assessment Programme 1998, p.312) that "Future monitoring should, therefore, include determination of total toxaphene (by NIMS) for comparison with past work as well as measurements of specific chlorobornane congeners."

Discussion and Conclusions

Quality assurance and quality control are essential elements of all research and monitoring programs. Ongoing interlaboratory comparison studies provide assurance on the quality and reliability of data being generated and on the comparability of measurement results between laboratories. Without this assurance of data quality and comparability, the value of measurement information upon which decisions are made will be lost. It is imperative that managers and decision makers of the NCP have a scientifically sound science base upon which to base their decisions.

The results of the two trace metal intercomparisons showed very positive results for the NCP measurement community. The sediment analyses were very comparable between participants, and the Z-scores were < 2 for all key heavy metals. At much lower concentrations, however, several NCP laboratories displayed considerable difficulty generating accurate and repeatable results, particularly for biotic samples. In fact, it could be said that some of the laboratories were "precisely inaccurate." On the other hand, the methylmercury results were excellent for the three tissue samples. Continued monitoring of trace metals and methylmercury analyses should continue on an annual basis, incorporating new test materials, such as caribou liver or kidney, in future studies.

In all intercomparisons, the participants are instructed to use their own routine methodology and analytical calibration standards. The recent OC/PCB study indicated that the calibration standards being used for NCP measurements are reliable and comparable. In future OC/PCB studies, an increasing progression of complexity in the test samples is planned, with a particular emphasis being placed on peak identification and detectability. Although the OC/PCB studies could be run every second year, toxaphene intercomparisons should be conducted on an annual basis.

The small repository of surplus check samples and standards with known target parameter concentrations will continue to be made available to NCP laboratories that experience difficulties with the study samples and require additional test material to evaluate the success of their corrective actions, and to those who wish to test new or modified methodologies. The development of reference materials as recommended by AMAP (Arctic

Monitoring and Assessment Programme 1998)) through the interlaboratory assessments should be considered in the future.

Current recommendations for the NCP interlaboratory QA program are:

- To conduct interlaboratory studies for organochlorines and PCBs every two years, including parameters such as the hexachlorocyclohexanes, chlordanes and PCB congeners at appropriate levels in both biotic and abiotic samples. Specific issues to be addressed are peak identification, resolution and detectability.
- To establish a series of toxaphene intercomparisons specific to the NCP research projects. The initial studies should establish the degree of comparability among the various NCP measurement laboratories analyzing different substrates by focussing on the data quality issues of peak identification and quantitation in standard solutions. Test samples should include injection-ready standards, abiotic extracts, lipid-free biota extracts, and eventually progress to more complex real matrix samples.
- To continue to evaluate the data quality of trace metal analyses, including those for mercury, methylmercury and selenium, in additional biotic samples.
- To address the quality and comparability of measurements for organotins, dioxins, PAHs, and other contaminants of emerging concern, as needed.

Thus, interlaboratory comparison exercises are, and will continue to be, the main activities of this QA program. Other activities, such as a maintaining an inventory of laboratories and their capabilities, and a list of external sources of QA support such as intercomparisons and reference materials, are also important to achieve the goals of this program. The combination of these efforts will not only provide for an evaluation of data quality within the NCP and to other international programs, but will also provide a means for continual improvement in the measurement process for the laboratories involved.

Expected Project Completion Date

The two surveys (Stokker and Gomes 1998, 1999) were completed in January 1999. As recommended by the QA Technical Sub-Committee, the laboratory survey will be updated annually prior to their review of the proposal for the next fiscal year's QA program. In addition, external QA programs will be reviewed on an ongoing basis in order to notify the appropriate NCP laboratories of the available intercomparisons suitable for their analyses.

The preliminary data review for the trace metals in sediment study was provided to the participants in March 1999 and for the biota study in December 1999. A report describing the full assessment of trace metal capabilities and comparability for both sediment and biota samples will be provided by August 2000.

The preliminary data review for the organochlorines and PCBs in biota study was provided to the participating laboratories in May 2000 and a final report will be issued by September 2000.

The overall performance of the NCP laboratories in these intercomparison studies is encouraging and will allow a stronger focus of attention to be placed on other areas of analysis in future exercises.

The assessment of analytical comparability for other contaminants and matrices is an ongoing process, with three additional intercomparisons being scheduled for 2000-2001. The test samples for Interlaboratory Study NCP-II-4 on the analysis of toxaphene in standard solutions and lipid-free burbot liver extract is being targeted for distribution in August 2000, as soon as test materials become available. Study NCP-II-5 on trace metals and methylmercury in muktuk, burbot liver and land-locked char muscle will also be delivered to participants in August 2000. Data summaries will be provided to the participants by December 2000. Interlaboratory Study NCP II-6 on the analysis of organotins is targeted for distribution in October or November 2000. Lastly, an assessment of the need for QA for stable lead isotope measurements will be made this year.

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